

A Neural Network-Based Application to Identify Cubic Structures in Multi Component Crystalline Materials using X-Ray Diffraction Data

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Summary

One of the crystalline materials structures is cubic. An experimental study has been done about developing a scheme to identify the cubic structure types in single or multi component materials. This scheme is using fingerprints created from the calculation of quadratic Miller indices ratios and matches it with the ratio of the $\sin^2\theta$ values from the diffracted data of material obtained by X-Ray Diffraction (XRD) method. These manual matching processes are complicated and sometimes are tedious because the diffracted data are complex and may have more than one fingerprint inside. This paper proposes an application of multi-layered back-propagation neural network in matching the fingerprints with the diffracted data of crystalline material to quickly and correctly identify its cubic structure component types.

Key words:

Artificial Neural Network, XRD Methods, Pattern Matching, Cubic Structure Fingerprints, Material Science.

Introduction

Crystallography has always been one of the most challenging research fields since eighteenth century. Following the discovery of X-ray for diffraction by Röntgen in 1895 [1], X-ray diffraction method has been applied to many different sub-area of crystallography such as identification of crystalline phases, qualitative and quantitative analysis of mixtures and minor constituents, distinction between crystalline and amorphous states, side-chain packing of protein structures, identification of crystalline material, etc. The evolution of crystallography research using X-ray diffraction method has always been tremendous and eventually touches the use of artificial intelligence techniques to fasten the process and increase the accuracy of the results.

This paper proposes an application of neural network into one of crystallography research area of identification of crystalline material. X-ray diffraction data interpretation for most crystalline materials is a very complex and

difficult task. This is due to the condition that different crystalline material may contain more than one cubic structures component type and after being diffracted using X-ray diffraction method, the diffracted data are complex. Hence, the data can be so ambiguous and not easy to track and understand. Neural networks can provide a fundamentally different approach to this crystalline material identification problem.

The ability of neural network to perform pattern recognition task has been proven to be good and useful in many physics and engineering applications. Attempts to apply neural network on crystallography research problems has been a great success so far. To name a few, Milik et al. [3] used it to evaluate the side-chain packing of protein structures, Berntson et al. [7] developed an application of a neural network to produce a high-throughput protein crystallography, Gallagher and Deacon [5] proposed an application of neural networks to classify mineralogical samples using X-ray spectra, Baker et al. [6] used it to recognize patterns in craniofacial X-ray image analysis and Su et al. [4] applied it to X-ray image Segmentation. The main intent of this paper is to examine further the ability of neural network to identify the cubic structures on multi component crystalline materials by performing fingerprints matching.

2. Cubic Structure Fingerprint Calculation

There are four cubic structures for crystalline material, the Simple Cubic (SC), Body Centered Cubic (BCC), Face Centered Cubic (FCC) and Diamond. In our previous work [2], a formula has been proposed to calculate the fingerprints for these four cubic structures. The formula utilizes the Miller index (h,k,l) [8]. The proposed formula can be written as follows:

$$\sin^2 \theta = \frac{\lambda^2 (h^2 + k^2 + l^2)}{4a^2} \quad (1)$$

Since the wavelength of the incoming X-ray (λ) and lattice constant (a) are both constants, we can eliminate these quantities from Eq. 1 and derive the ratio of two $\sin^2\theta$ as follows:

$$R_{m,n} = \frac{\sin^2 \theta_m}{\sin^2 \theta_n} = \frac{h_m^2 + k_m^2 + l_m^2}{h_n^2 + k_n^2 + l_n^2} \quad (2)$$

where θ_m and θ_n are the diffracting angles for two peak associated with the diffracting planes $\{h_m, k_m, l_m\}$ and $\{h_n, k_n, l_n\}$ respectively for ratio $R_{m,n}$.

The fingerprint for each crystalline material's cubic structure is calculated by taking 10 peaks from the X-ray diffraction data and calculates the two peaks combinations from that 10 peaks. That is, each fingerprint is actually contains 45 values of the quadratic sinus ratio from Eq. 2 since C_2^{10} equal to 45. Table 1 and Table 2 show the fingerprint for Face Centered Cubic (FCC) and Diamond respectively.

Table 1: Face Centered Cubic (FCC) Fingerprint

$R_{1,2} = 0.750$	$R_{2,3} = 0.500$	$R_{3,5} = 0.666$	$R_{4,8} = 0.550$	$R_{6,8} = 0.800$
$R_{1,3} = 0.375$	$R_{2,4} = 0.364$	$R_{3,6} = 0.500$	$R_{1,9} = 0.458$	$R_{6,9} = 0.666$
$R_{1,4} = 0.273$	$R_{2,5} = 0.333$	$R_{3,7} = 0.421$	$R_{1,10} = 0.407$	$R_{6,10} = 0.593$
$R_{1,5} = 0.250$	$R_{2,6} = 0.250$	$R_{3,8} = 0.400$	$R_{5,6} = 0.750$	$R_{7,8} = 0.950$
$R_{1,6} = 0.187$	$R_{2,7} = 0.210$	$R_{3,9} = 0.333$	$R_{5,7} = 0.632$	$R_{7,9} = 0.792$
$R_{1,7} = 0.158$	$R_{2,8} = 0.200$	$R_{3,10} = 0.296$	$R_{5,8} = 0.600$	$R_{7,10} = 0.704$
$R_{1,8} = 0.150$	$R_{2,9} = 0.166$	$R_{4,5} = 0.916$	$R_{5,9} = 0.500$	$R_{8,9} = 0.833$
$R_{1,9} = 0.125$	$R_{2,10} = 0.148$	$R_{4,6} = 0.687$	$R_{5,10} = 0.444$	$R_{8,10} = 0.741$
$R_{1,10} = 0.111$	$R_{3,4} = 0.727$	$R_{4,7} = 0.579$	$R_{6,7} = 0.842$	$R_{9,10} = 0.888$

Table 2: Diamond Fingerprint

$R_{1,2} = 0.375$	$R_{2,3} = 0.727$	$R_{3,5} = 0.579$	$R_{4,8} = 0.500$	$R_{6,8} = 0.750$
$R_{1,3} = 0.273$	$R_{2,4} = 0.500$	$R_{3,6} = 0.458$	$R_{1,9} = 0.457$	$R_{6,9} = 0.686$
$R_{1,4} = 0.187$	$R_{2,5} = 0.421$	$R_{3,7} = 0.407$	$R_{1,10} = 0.400$	$R_{6,10} = 0.600$
$R_{1,5} = 0.158$	$R_{2,6} = 0.333$	$R_{3,8} = 0.344$	$R_{5,6} = 0.792$	$R_{7,8} = 0.844$
$R_{1,6} = 0.125$	$R_{2,7} = 0.296$	$R_{3,9} = 0.314$	$R_{5,7} = 0.704$	$R_{7,9} = 0.771$
$R_{1,7} = 0.111$	$R_{2,8} = 0.250$	$R_{3,10} = 0.275$	$R_{5,8} = 0.594$	$R_{7,10} = 0.675$
$R_{1,8} = 0.094$	$R_{2,9} = 0.228$	$R_{4,5} = 0.842$	$R_{5,9} = 0.543$	$R_{8,9} = 0.914$
$R_{1,9} = 0.086$	$R_{2,10} = 0.200$	$R_{4,6} = 0.666$	$R_{5,10} = 0.457$	$R_{8,10} = 0.800$
$R_{1,10} = 0.075$	$R_{3,4} = 0.687$	$R_{4,7} = 0.593$	$R_{6,7} = 0.889$	$R_{9,10} = 0.875$

In this paper we only use these two fingerprints on our experiments due to limited availability of the crystalline materials of other cubic structure component types. The rest of the fingerprint details are described at [2].

3. Back-Propagation Neural Networks

Fig. 1 depicts the architecture of the proposed application. The main module is parted into two parts. The first part is the preprocessing and training part. This part will train the neural network for each fingerprint and save the resulted network preference.

The second part is the neural network itself. In this application we use the multi-layered back-propagation neural network. It is a central and the most commonly used tool on modeling and classification problems. Input-output pairs are presented to the network, and weights are adjusted to minimize the error between the network output and the actual value. Our neural network consists of three layers of neurons, an input layer, two hidden layers and an output layer.

The standard iterative gradient algorithm for back-propagation training is used by the proposed application. It is designed to minimize the Mean Square Error (MSE) between the predicted output and the desired output. The training algorithm used is summarized as follows:

1. Initialize weights and threshold values: set all weights and threshold to small random values.
2. Present input and desired output: present a continuous valued input vector X_1, X_2, \dots, X_n , and specify the desired outputs O_1, O_2, \dots, O_n .
3. Compute the output of each node in both hidden layer:

$$h_j = f\left(\sum_{i=1}^n W_{ij} X_i - \theta_j\right) \quad (3)$$

4. Compute the output of each node in the output layer:

$$O_k = f\left(\sum_{i=1}^m W_{ki} X_i - \theta_k\right) \quad (4)$$

and

$$f(x) = \frac{1}{1 + e^{-x}} \quad (5)$$

and

$$\delta_k = O_k(1 - O_k)(Y - O_k) \quad (6)$$

5. Compute both hidden layer error:

$$\delta_j = h_j(1 - h_j) \sum_{k=1}^m \delta_k W_{kj} \quad (7)$$

6. Compute the output layer error between the target and the observed output using Eq. 6.

7. Re-compute both hidden layer error using Eq. 7.

8. Adjust the weights and thresholds in the output layer:

$$W_{kj}(t+1) = W_{kj}(t) + \alpha\delta_k h_j + \eta(W_{kj}(t) - W_{kj}(t-1)) \quad (8)$$

and

$$\theta_k(t+1) = \theta_k(t) + \alpha\delta_k \quad (9)$$

9. Adjust the weights and thresholds in both hidden layer:

$$W_{ji}(t+1) = W_{ji}(t) + \alpha\delta_j h_i + \eta(W_{ji}(t) - W_{ji}(t-1)) \quad (10)$$

and

$$\theta_j(t+1) = \theta_j(t) + \alpha\delta_j \quad (11)$$

10. Repeat steps 2 to 9 on the all pattern pairs until the output layer error is within the specified tolerance for each pattern and for each neuron.

4. Experiments and Results

The experiments are carried out using Philips' X-Ray device diffractometer control PW1710. The device was using PW1729 series of X-Ray generator, anode Cu tube, and PCAPD (PW1877) software version 3 integrated with our proposed application installed. In the overall experiments, we use the same tube voltage and also the same tube current, i.e. 30kV and 20mA respectively.

Three samples of crystalline material are used in the experiment. The samples are Aluminium (Al) and Silicon (Si) materials. For the purpose to have a multi component crystalline material, we mix Al sample and Si sample and create the third sample which is a mixture of Al and Si.

The neural network structure presented on Fig. 1 was chosen after we train several neural network structures using the fingerprints on Table 1 and Table 2. The result of the training process is presented on Table 3.

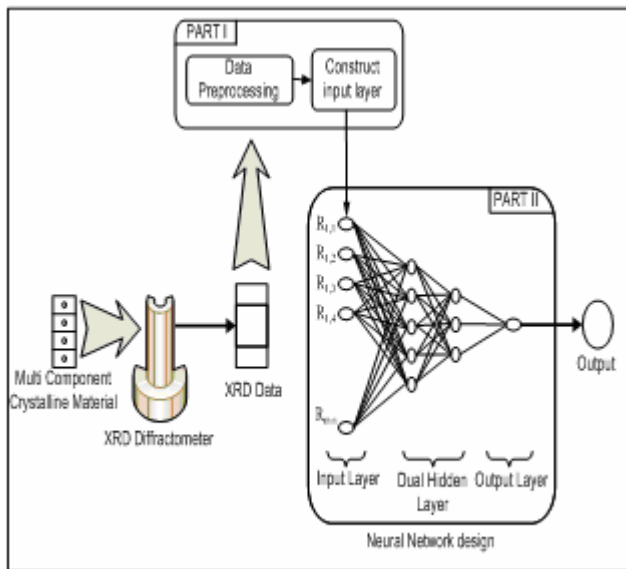


Fig. 1 The Proposed Application Architecture with Back-Propagation Neural Network

The proposed application will behave as an integrated application with the XRD Diffractometer software to receive the diffracted data directly for faster and reliable processing. At the end, a report will be generated which contains the cubic structure component type detected on that particular crystalline material.

Table 3: Neural Network Training Results

<i>Fingerprint</i>	<i>MSE</i>	<i># of Nodes on Hidden Layer</i>
Aluminium (Al)	4.80	5
	4.34	6
	4.35	7
	4.54	8
	4.30	9
	4.32	10
	4.59	11
Silicon (Si)	4.36	12
	4.86	5
	4.59	6
	4.61	7
	4.44	8
	4.67	9
	4.71	10
	4.60	11
4.66	12	

The Silicon (Si) training results show that the lowest Mean Square Error (MSE) is 4.44 with 8 nodes in hidden layer while Aluminium (Al) training results show that the lowest MSE is 4.30 with 9 nodes in the hidden layer. Hence we construct our neural network with 8 nodes in the hidden layer, 5 in the first hidden layer and 3 in the second hidden layer.

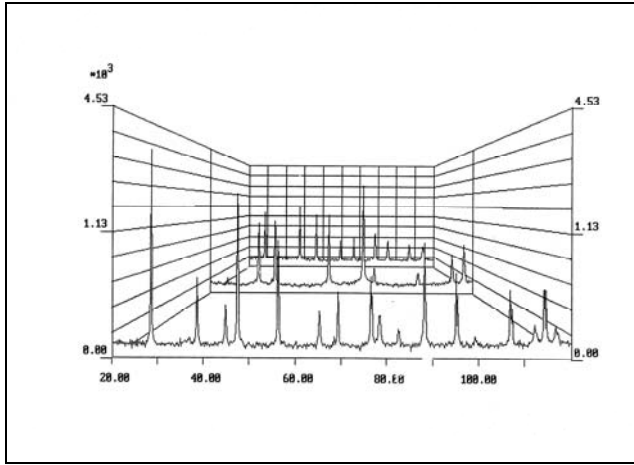


Fig. 2 X-Ray Diffraction Pattern for the Mixture Sample of Aluminium and Silicon

Fig 2 visualizes the results obtained from X-ray diffraction method after we load the samples of the crystalline material into the X-ray diffractometer. The front pattern is the diffraction data result for the mixture sample of Aluminium and Silicon, the middle pattern is the diffraction data result for Aluminium sample material, and the last one on the back is the diffraction data result for Silicon sample material.

From the resulted diffraction data for the mixture sample, we can see that it has 17 peaks and therefore we applied the combination of two peaks from that 17 peaks (C_2^{17}) and we have the ratio $R_{m,n}$ values shown on Table 5. The same calculation was performed on the resulted diffraction data for the original Aluminium and Silicon samples and their ratio $R_{m,n}$ are shown at Table 4 and Table 6 respectively. Next is our proposed application will pick up these results and feed it into the previously trained neural network.

Table 4: Diffracted Data for the Sample of Aluminium

$R_{1,2} = 0.750$	$R_{2,3} = 0.499$	$R_{3,5} = 0.666$	$R_{4,8} = 0.550$	$R_{6,9} = 0.666$
$R_{1,3} = 0.374$	$R_{2,4} = 0.363$	$R_{3,6} = 0.501$	$R_{1,9} = 0.458$	$R_{7,8} = 0.950$
$R_{1,4} = 0.272$	$R_{2,5} = 0.333$	$R_{3,7} = 0.421$	$R_{5,6} = 0.751$	$R_{7,9} = 0.791$
$R_{1,5} = 0.249$	$R_{2,6} = 0.250$	$R_{3,8} = 0.400$	$R_{5,7} = 0.632$	$R_{8,9} = 0.833$
$R_{1,6} = 0.187$	$R_{2,7} = 0.210$	$R_{3,9} = 0.333$	$R_{5,8} = 0.600$	
$R_{1,7} = 0.158$	$R_{2,8} = 0.200$	$R_{1,5} = 0.916$	$R_{5,9} = 0.500$	
$R_{1,8} = 0.150$	$R_{2,9} = 0.166$	$R_{1,6} = 0.688$	$R_{6,7} = 0.842$	
$R_{1,9} = 0.125$	$R_{3,4} = 0.727$	$R_{1,7} = 0.579$	$R_{6,8} = 0.799$	

Table 5: Diffracted Data for the Mixture Sample of Aluminium and Silicon

$R_{1,2} = 0.558$	$R_{2,14} = 0.168$	$R_{4,14} = 0.250$	$R_{7,8} = 0.842$	$R_{10,11} = 0.898$
$R_{1,3} = 0.419$	$R_{2,15} = 0.157$	$R_{4,15} = 0.233$	$R_{7,9} = 0.808$	$R_{10,12} = 0.799$
$R_{1,4} = 0.375$	$R_{2,16} = 0.154$	$R_{4,16} = 0.228$	$R_{7,10} = 0.741$	$R_{10,13} = 0.749$
$R_{1,5} = 0.273$	$R_{2,17} = 0.150$	$R_{4,17} = 0.222$	$R_{7,11} = 0.666$	$R_{10,14} = 0.673$
$R_{1,6} = 0.209$	$R_{3,4} = 0.897$	$R_{5,6} = 0.764$	$R_{7,12} = 0.593$	$R_{10,15} = 0.631$
$R_{1,7} = 0.188$	$R_{3,5} = 0.652$	$R_{5,7} = 0.687$	$R_{7,13} = 0.554$	$R_{10,16} = 0.615$
$R_{1,8} = 0.158$	$R_{3,6} = 0.500$	$R_{5,8} = 0.579$	$R_{7,14} = 0.500$	$R_{10,17} = 0.600$
$R_{1,9} = 0.152$	$R_{3,7} = 0.448$	$R_{5,9} = 0.555$	$R_{7,15} = 0.467$	$R_{11,12} = 0.889$
$R_{1,10} = 0.139$	$R_{3,8} = 0.377$	$R_{5,10} = 0.509$	$R_{7,16} = 0.456$	$R_{11,13} = 0.832$
$R_{1,11} = 0.125$	$R_{3,9} = 0.363$	$R_{5,11} = 0.458$	$R_{7,17} = 0.444$	$R_{11,14} = 0.750$
$R_{1,12} = 0.111$	$R_{3,10} = 0.333$	$R_{5,12} = 0.407$	$R_{8,9} = 0.960$	$R_{11,15} = 0.701$
$R_{1,13} = 0.104$	$R_{3,11} = 0.299$	$R_{5,13} = 0.381$	$R_{8,10} = 0.880$	$R_{11,16} = 0.686$
$R_{1,14} = 0.093$	$R_{3,12} = 0.265$	$R_{5,14} = 0.343$	$R_{8,11} = 0.792$	$R_{11,17} = 0.666$
$R_{1,15} = 0.087$	$R_{3,13} = 0.250$	$R_{5,15} = 0.321$	$R_{8,12} = 0.704$	$R_{12,13} = 0.936$
$R_{1,16} = 0.085$	$R_{3,14} = 0.224$	$R_{5,16} = 0.313$	$R_{8,13} = 0.659$	$R_{12,14} = 0.844$
$R_{1,17} = 0.083$	$R_{3,15} = 0.210$	$R_{5,17} = 0.305$	$R_{8,14} = 0.593$	$R_{12,15} = 0.788$
$R_{2,3} = 0.750$	$R_{3,16} = 0.204$	$R_{6,7} = 0.899$	$R_{8,15} = 0.555$	$R_{12,16} = 0.770$
$R_{2,4} = 0.673$	$R_{3,17} = 0.200$	$R_{6,8} = 0.757$	$R_{8,16} = 0.542$	$R_{12,17} = 0.749$
$R_{2,5} = 0.490$	$R_{4,5} = 0.727$	$R_{6,9} = 0.727$	$R_{8,17} = 0.527$	$R_{13,14} = 0.899$
$R_{2,6} = 0.375$	$R_{4,6} = 0.556$	$R_{6,10} = 0.666$	$R_{9,10} = 0.916$	$R_{13,15} = 0.842$
$R_{2,7} = 0.336$	$R_{4,7} = 0.500$	$R_{6,11} = 0.599$	$R_{9,11} = 0.824$	$R_{13,16} = 0.822$
$R_{2,8} = 0.283$	$R_{4,8} = 0.421$	$R_{6,12} = 0.532$	$R_{9,12} = 0.732$	$R_{13,17} = 0.800$
$R_{2,9} = 0.272$	$R_{4,9} = 0.404$	$R_{6,13} = 0.500$	$R_{9,13} = 0.686$	$R_{14,17} = 0.936$
$R_{2,10} = 0.250$	$R_{4,10} = 0.370$	$R_{6,14} = 0.448$	$R_{9,14} = 0.617$	$R_{15,16} = 0.914$
$R_{2,11} = 0.224$	$R_{4,11} = 0.333$	$R_{6,15} = 0.421$	$R_{9,15} = 0.578$	$R_{15,17} = 0.889$
$R_{2,12} = 0.199$	$R_{4,12} = 0.296$	$R_{6,16} = 0.410$	$R_{9,16} = 0.564$	$R_{16,17} = 0.976$
$R_{2,13} = 0.187$	$R_{4,13} = 0.277$	$R_{6,17} = 0.400$	$R_{9,17} = 0.550$	$R_{9,10} = 0.950$
				$R_{9,10} = 0.973$

Table 6: Diffracted Data for the Sample of Silicon

$R_{1,2} = 0.374$	$R_{2,3} = 0.727$	$R_{3,5} = 0.579$	$R_{4,8} = 0.500$	$R_{6,8} = 0.751$
$R_{1,3} = 0.272$	$R_{2,4} = 0.500$	$R_{3,6} = 0.458$	$R_{1,9} = 0.458$	$R_{6,9} = 0.687$
$R_{1,4} = 0.187$	$R_{2,5} = 0.421$	$R_{3,7} = 0.407$	$R_{1,10} = 0.400$	$R_{6,10} = 0.600$
$R_{1,5} = 0.157$	$R_{2,6} = 0.333$	$R_{3,8} = 0.344$	$R_{5,6} = 0.792$	$R_{7,8} = 0.845$
$R_{1,6} = 0.125$	$R_{2,7} = 0.296$	$R_{3,9} = 0.315$	$R_{5,7} = 0.704$	$R_{7,9} = 0.772$
$R_{1,7} = 0.111$	$R_{2,8} = 0.250$	$R_{3,10} = 0.275$	$R_{5,8} = 0.595$	$R_{7,10} = 0.675$
$R_{1,8} = 0.094$	$R_{2,9} = 0.229$	$R_{4,5} = 0.842$	$R_{5,9} = 0.544$	$R_{8,9} = 0.914$
$R_{1,9} = 0.085$	$R_{2,10} = 0.200$	$R_{4,6} = 0.666$	$R_{5,10} = 0.475$	$R_{8,10} = 0.799$
$R_{1,10} = 0.075$	$R_{3,4} = 0.687$	$R_{4,7} = 0.593$	$R_{6,7} = 0.889$	$R_{9,10} = 0.875$

Table 7 depicts the results of the proposed neural network-based application. It is clear that the neural network successfully recognizes the fingerprint inside the diffraction data of each sample. For the mixture sample, the results show that it has two cubic structure types, Face Centered Cubic (FCC) and Diamond. The detection accuracy was 86% for FCC and 84% for Diamond. This result is supported by the results of the original Aluminium and Silicon samples. The Aluminium sample was detected to have a FCC type with detection accuracy of 92% while the Silicon sample was detected to have a Diamond type with detection accuracy of 91%.

Table 7: Experiments Results

<i>Samples</i>	<i>Detected Type</i>	<i>Detection Accuracy</i>
Aluminium (Al)	FCC	92%
Silicon (Si)	Diamond	91%
Mixture of Al and Si	FCC	86%
	Diamond	84%

Lastly, the following was the report generated by the proposed application for experiment with the mixture sample. The reports for the other two samples are basically the same.

```

SampleName: Al&Si
DiffractionDataFile: File-5-0565
DetectedType: FCC, Diamond
Accuracy: FCC(86%), Diamond(84%)
GeneratedOn: 15:17:42-14Feb2007
    
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5. Conclusion

In this paper we propose an application that uses multi layer back-propagation neural network to identify cubic structures on multi-component crystalline material. The use of neural network proves efficient and faster in recognizing the fingerprints inside the diffracted crystalline material data.

This work is a good start towards the use of neural network on crystalline material identification research. While this paper is being written, we are in the process of extending our experiments to include more crystalline materials and extending the time duration in performing the X-ray diffraction process in order to have more convincing outputs. Our future direction includes enhancing the neural network structure and applying rule-based fuzzy neural network to increase the accuracy.

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