IDENTIFICATION OF CRYSTALLINE STRUCTURES USING MOSSBAUER PARAMETERS AND ARTIFICIAL NEURAL NETWORK


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Mössbauer spectroscopy is a useful technique for characterizing the valencies, electronic and magnetic states, coordination symmetries and site occupations of Fe cations. The Mössbauer parameters of Isomer Shift (I.S.) and Quadrupole Splitting (Q.S.) are useful to distinguish paramagnetic ferrous and ferric ions in several substances, while the internal magnetic field provides information on the crystallinity. A correlation is being sought between Mössbauer parameters and several structure properties of some iron-containing minerals using Artificial Neural Networks (ANN). Distinct regions of crystalline structures are defined when any two parameters are plotted, but in several cases superposition of these regions leads to erroneous conclusions. We have tried to eliminate this difficulty by using convenient axes. These axes form n-dimensional vectors as input to our ANN. In recent years ANN has shown to be a powerful technique to solve problems as pattern recognition, optimization, preview ups and downs in stock market, automatic control and identification of a mineral from a Mössbauer spectrum or Mössbauer data bank. Using ANN we have been successful in identification of crystalline structures from plots of Mössbauer spectral parameters of I.S., Q.S., and structure properties of mean metal-oxygen distance in coordination site. Results using ANN in identification of crystalline structures using Mössbauer parameters of I.S., Q.S., and polyhedral volume of a coordination site are presented.

A correlation between isomer shift (I.S.) and quadrupole splitting (Q.S.) for a series of mono-substituted ferrocyanides was attempted by Brant. Brady et al. observed that the data then available showed no general correlation between I.S. and Q.S. in high spin ferrous compounds. For anhydrous ferrous halides, ferrous chloride hydrates and ferrous iodide hydrates approximately linear relationships have been reported. The Q.S. data employed in the correlations were limited to compounds in the paramagnetic phase at temperatures sufficiently low that the splitting attained a constant value. All the compounds contained octahedrally coordinated Fe\(^{2+}\) and the observed correlations indicated minimal contributions to the Q.S. in these compounds from the lattice spin orbit coupling. Jorgensen proposed that linear correlations result from similar effects on both I.S. and Q.S. that are produced by differences in the central field covalency. Burger and Bancroft et al. tried to correlate this by partial values of I.S. and Q.S. There have been attempts to correlate I.S. and Q.S. to obtain additional information. Burns reported these parameters graphically in pairs making it in 2-dimensional graphics. In some cases (orthopyroxene I.S.=1.12 mm/s and Q.S.=1.90 mm/s, octahedral; and pigeonite I.S.=1.12 mm/s and Q.S.=1.96 mm/s, 6-7 coordination) there are superposition of clusters that made impossible the precise determination of their crystalline structures using only I.S. and Q.S. To identify correctly it was necessary to make various 2-dimensional graphics.

We have attempted, in the present study, to solve this limitation by utilizing 3 or more dimensional graphics. The learning and classification process of Artificial Neural Network (ANN) perform this task with quickness and precision. Interest in ANN has been growing rapidly over the last few years and researchers from diverse fields are intrigued by the potential offered by this technology. This interest has been inspired by both theoretical and applicational successes. Now it is
possible to apply computation to make machines that learn and remember in ways that bear a striking resemblance to human mental processes. Artificial neurons learn from experience, generalize from previous examples to new ones, and abstract essential characteristics from inputs containing irrelevant data. Once the networks are trained, response can be insensitive to minor variations in its inputs. The ability to see through noise and distortion to the pattern that lies within is vital for the pattern recognition in a real world environment. An ANN can be trained with sequence of a set of inputs, and after adequate training it can produce something that has never been seen before. It will have the ability to extract an ideal from imperfect inputs. Minsky and Papert first reported single layer networks (perceptron) and their incapabilities. There have been some very impressive demonstrations of ANN capabilities using backpropagation networks, discovered independently by Werbos and Parker, and Rumelhart et al. that provide a systematic means for training multilayer networks. ANN has been used for identification of substances using Mössbauer spectra and parameters. Neural computing, new developments, theory and practice have been reported by many workers.

The present study involves the training of hybrid networks called counterpropagation with the Mössbauer parameters of isomer shift and quadrupole splitting from the literature, and another training with these parameters as well as with the values of polyhedral volume of a coordination site. Adequately trained ANN could successfully identify, from new values of these parameters, the crystalline structure listed in Table II.

Theory

An artificial neural network is an information-processing system that has certain performance characteristics in common with biological neural networks. ANN has been developed as generalizations of mathematical models of biological neuron, based on the following assumptions:

1. Information processing occurs at many simple elements called neurons, and the signals are passed between neurons over connection links.
2. Each connection link has an associated weight, which, in a typical neural network, multiplies the signal transmitted.
3. Each neuron applies an activation function (usually nonlinear) to its input (sum of weighted input signal) to determine its output signal.

In an artificial neuron (Fig. 1b) input branch of dendrites communicates with the signal from other neurons. The neuron does simple processing and gives numerical result to its output branch (axon), which is represented as scalar product

\[ S = \sum_{i=1}^{n} W_i X_i = WX \]

here \( W_i \) and \( X_i \) are the coordinates of the weight vector \( W \) and the input vector \( X \), respectively.

Figure 1a: A biological neuron.

Figure 1b: An artificial neuron.

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