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Artifficial Neural Network Modeling for AC conductivity Behaviour of PVA/acid salt Polymer Electrolyte

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Abstract— Levenberg-Marquardt algorithm (LM) and neural networks (NN) are combined to study the electrical properties of PVA /Acid Salt Polymer Electrolytes $(PVA)_{(1-x)}(MgBr_2)_{x/2}(H_3PO_4)_{x/2}$. The obtained function from NN model calculates and simulates the relation between the AC conductivity and the frequency for PVA/acid salt at different temperatures. The simulation results from NN-based model are compared with the experimental data. The obtained function of NN model has proven matching better for the experimental data. The results show that NNs are able to produce accurate results of the electrical properties for PVA/acid salt polymer electrolytes.

Index Terms— Artificial Neural Network (ANN), AC conductivity, Acid Salt, Polymer, Electrolytes, Levenberg-Marquardt algorithm and PVA.

1 Introduction

Solid Polymer Electrolytes (SPEs) in the highly ionic conducting, electrochemical stability and in the flexible film form have been attracting great deal of attentions in the recent years because of their potential technological application relevance's in developing all-solid-state electrochemical devices viz. batteries, fuel cells, super capacitors, ECDs etc. [1, 2].

Extensive studies have been undertaken to investigate ion conduction behavior on polymer materials. Impedance spectroscopy is employed to establish the conduction mechanism observing the contribution of the polymeric chain mobility and carrier generation processes. One of the most characteristic features of electrical conduction in disordered solid systems is the dispersion of conductivity with frequency. In the low frequency regime, almost found to be frequency independent - the plateau value - and it is equal to true dc conductivity. While in the high frequency region, closer to the relaxation times, the mobility of the charge carriers is high and hence, the conductivity increases with frequency and varying approximately as a power of frequency [3–6]. Different approaches have been used for the interpretation of the conductivity performance of these disordered composites.

All these models can be broadly categorized in two categories, namely microscopic and macroscopic models. The microscopic model [7] assumes disorder on atomic length scale and the conductivity is due to either hopping or tunneling through the localized states. Whereas the macroscopic model [8] assumes disorder on length scales large enough that a local conductivity may be defined and the conductivity is explained on the basis of either effective medium approximation (EMA) or percolation path approximation (PPA).

In this connection, the neural network (NN)[9-11] has been studied and designed to study the AC conductivity (σ) and frequency for PVA/acid salt polymer electrolytes $(PVA)_{(1-x)}(MgBr_2)_{x/2}(H_3PO_4)_{x/2}$. AC conductivity is calculated by minimizing the differences between measured and model-generated results. The Levenberg-Marquardt algorithm is used to a measure of the quality of the match between the experimental data and model calculated. Modeling tools play an important role in Solid State Physics. Neural network approaches provide an effective tool [12, 13] for such modeling.

In the present work, we illustrate the experimental technique to prepare the PVA/acid salt electrolytes. Following sections provide a brief introduction of ANN to model the relationship between AC conductivity (σ) and frequency for PVA/acid salt and discuss the results.

2 Experiment Details

The preparation of polymer complexes has been described elsewhere [2]. The polymer films were kept in desiccators for further drying. In order to study the amorphous character of PVA complexes with different weight percentages of $MgBr_2$ and H_3PO_4, the XRD patterns of the samples were recorded at room temperature with a Philips X'Pert instrument, which employs a CuKa X-radiation.
To study the ionic conductivity of the samples, impedance spectroscopy was performed using a HIOKI 3532 programmable automatic LCR bridge interfaced to a computer for data acquisition. The study was carried out in the frequency range 100 Hz to 100 kHz. The thin polymer electrolyte films were sandwiched between two stainless steel disk electrodes, which acted as a blocking electrode for ions. The temperature-dependent ionic conductivity was performed in the tempera-

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ture range between 30 and 150 °C. The conductivity, σ, of each sample was calculated using equation $\sigma = t/(R_b A)$, where t is the thickness of the film, R_b is the bulk impedance and A is the area of the cross-section of the film. The value of R_b was confirmed by comparing with the $1/R_b$ value obtained from a complex admittance graph.

3 Neural Network Model

Soft computing is a practical alternative for solving complex problems through the use of human expertise and a prior knowledge about the problem in hand. Neural network is made up of many simple and highly interconnected computational elements [14-16], called artificial neurons or nodes.

The net input into j^{th} layer node [in (j)] is equal to the sum of weighted outputs from the prior i^{th} layer [out (i)]

$$in[j] = \sum w_{ij}\ out\ [i] \qquad (1)$$

where, w_{ij} is the weight factor.

The neurons of hidden layers and the weight factors of the links between them play a critical role during the learning process. A simple neuron structure is shown in the fig. (1)

The transfer function (φ) of processing nodes is used to determine the output value of the node based on the total net input from nodes in prior layer. The neuron has a bias (b_k), which is summed with the weighted inputs to form the net input to transfer function (φ). Multilayer network, shown in fig(2), consists of one or more layers of neurons, called hidden layer, between input and output layer. The network is called fully connected when every neuron in one layer is connected to every neuron of the next layer.

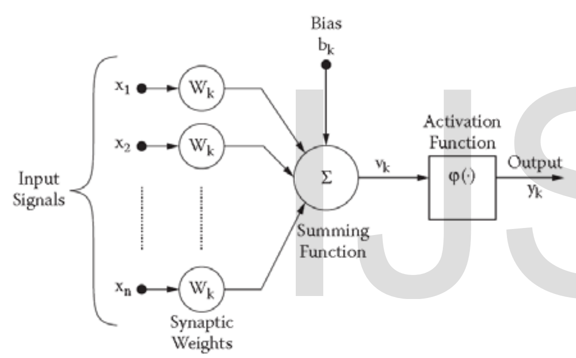

Fig. 1. Neuron structure.

The proposed ANN model was trained using Levenberg-Marquardt (LM) optimization technique [17-19].

This optimization technique is more powerful than the conventional gradient decent techniques. The adjustment for the weights (Δw) is done by the following equation

$$\Delta w = (j^T j + \mu I)^{-1} j^T e \qquad (2)$$

where j is the Jacobin matrix of derivatives of each error with respect to each weight, j^T is the transposed matrix of j, (I) is the identity matrix that has the same dimensions as those of $j^T j$, (μ) is a scalar changed adaptively by the algorithm and e is an error vector. Also Δw is a measure for the rate of learning of the network.

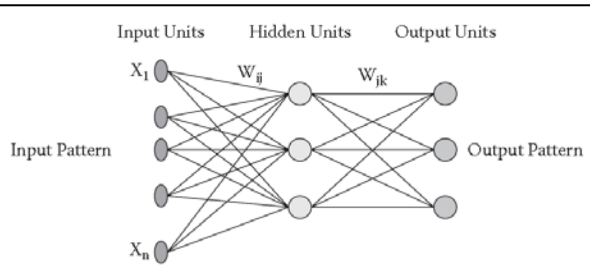

Fig. 2. General architecture of a multilayer perceptron.

4 Results

The proposed neural network model for AC conductivity (σ) have two inputs: frequency (f) and temperature (T), one output (σ) and three hidden layers which consists of 10, 10 and 7 neurons respectively as shown in fig. (3).

The transfer function where chosen to be a tan sigmoid function $[(e^{net} - e^{-net})/(e^{n} + e^{-n})]$ for the hidden layer and a pure line function (linear function) for the output layer. Using this input-output arrangement and connecting weights (Iw, Lw) are trained using LM for the given experimental data as described in sec 2. It is interesting to note that the training reached a zero sum square error which means that an exact fitting for the experimental data as seen in fig. (4). Appendix shows the final NN weights after training of 2000 Epochs and stopped after

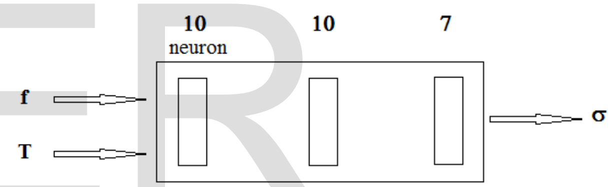

Fig. 3. Representation of ANN Modeling for AC conductivity (σ).

19^{th} iteration. Also in appendix the obtained function for AC conductivity (σ) using NN model which calculate the relation between AC conductivity and the frequency for PAV/acid salt polymer electrolytes $(PVA)_x(MgBr_2)_{x/2}\,(H_3PO4)_{x/2}$.

Fig (4) illustrates the simulation results of σ as a function of frequency at given temperature. The proposed σ based ANN model was tested after training on:

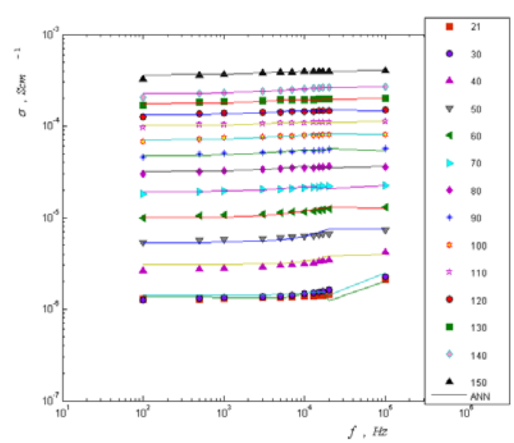

Fig. 4. Representation of ANN Modeling for AC Conductivity (σ).

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1. 80% data sets used in training (to assure the simulation capability of the proposed ANN).
2. 20% data sets not used in the training (to assure the prediction capability of the proposed ANN).

Results of σ as in fig.(4) based ANN model showed good fitting to the experimental data. This gives the ANN the provsion of wide usage in modeling of solid state physics.

Appendix A:

The equation which describes the relation between the AC conductivity and frequency is given by:

σ = *Pureline [{net.LW (4,3). tan sigmoid {net . LW(3,2). tan sigmoid {net . LW(2,1). tan sigmoid {net.IW(1,1)β + net.b(1)} + net.b(2)} + net.b(3)} + net.b(4)}]*

Where, *β is the input which is (Temp, f(Hz)).*

net.LW (4,3) linked weight between the third hidden layer and the output layer.

net.LW (3,2) linked weight between the third hidden layer and the second hidden layer .

net.LW (2,1) linked weights between the first and the second hidden layer.

net.IW (1,1)linked weights between the input layer and the first hidden layer.

net.b (1) is the bias of the first hidden layer.

net.b (2) is the bias of the second hidden layer.

net.b (3) is the bias of the third hidden layer.

net.b (4) is the bias of the output layer.

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