Indian Journal of Pure & Applied Physics
Vol. 58, June 2020, pp. 486-493

Computational studies for the effective electrical conductivity of Copper powder filled LDPE/LLDPE composites

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Received 1 January 2020; accepted 20 April 2020

The effective electrical conductivity (EEC) of low density polyethylene (LDPE) and linear low density polyethylene (LLDPE) polymer composites filled with copper has been studied. The nonlinear behavior has been observed for effective electrical conductivity versus filler content. Several approaches have been described to predict the electrical conductivities of polymer composites. EEC is described by artificial neural network (ANN) and it demonstrates the accurate match of experimental data for EEC with different training functions (TRAINOSS, TRAINLM, TRAINBR, TRAINSCG, TRAINBFG, and TRAINRP). The ANN approach satisfied the experimental data for EEC of polymer composites reasonably well. The complex structure encountered in LDPE/Cu and LLDPE/Cu, along with the difference in the EEC of the components, make it difficult to estimate the EEC exactly. This is the reason for which artificial neural network has been employed here. By using ANN approach experimental results indicate that EEC of polymer composites increases with increasing filler content at the same concentration.

Keywords: Effective electrical conductivity, Artificial neural network, Training functions, Volume fraction

1 Introduction

Electrical conductive polymer composite is formed by adding insulating polymer matrix with conducting filler like copper particle, metal particle and carbon particle[1,2]. Conducting polymer composite has many remarkable properties because of the resistivity variation with mechanical, electrical and thermal conductivity. Conductive polymer composite materials have many applications in engineering, shielding, switching, sensors or self-regulated heating[2]. Polymer contain thermal and electrical insulating behavior like epoxy, rubber etc. These composites have many practical applications in electrical shielding, electrical heating and in analytical devices. The property of conducting phase is adjustable and when the volume fraction of conducting filler is low, the resistivity of its components is close to insulating material. The effective electrical conductivity can be improved by increasing the volume fraction of conductive filler. The schematic of artificial neural Network approach is explained in Fig. 1.

Polymer composite filled with metal has electrical character which is close to the property of metal. The metallic property of these composites depends upon many factors like electrical and physical characteristics[3]. The transfer of heat flow and electrical charge determine the EEC of polymer. The behavior of conductive polymer composite also depends upon the shape of particles and spatial distribution with polymer matrix. In past few years some approach have been developed to study the effective properties of two phase composite materials[4] but the investigations on the size and shape dependence of EEC of composites are still limited. To explain such phenomenon, we built a theoretical approach for polymer composites comprising copper particles. Theoretical prediction is not only useful for the purpose of analysis and optimization of the performance of composite material but also for the developments of new designs. This has been become possible after theoretical prediction of EEC for multi-phase composite materials.

ANNs are electronic models works on principle of brain's neural structure. This brain modeling plays a less technical role in developing the machine solutions. The ANN is novel way to compute the data and provide interesting graceful degradation during

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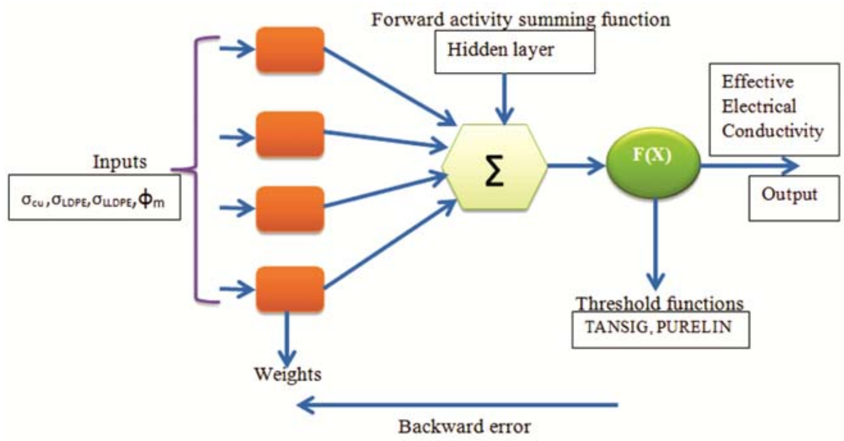

Fig. 1 — Artificial neural network approach.

system overload[7]. Nowadays, the biologically inspired methods are good tools of computing. Researchers have reported that animal brains are capable of some functions which are limited by computers[8,9]. But computers have not capable recognizing simple patterns and much less efficiency those patterns of the past into actions of the future[7]. Computers performs mathematics with good efficiency but unable to map simple pattern of the past into future actions.

Biological research gives understanding of the natural thinking mechanism. Present study validates that brains store information in the forms of patterns. Few patterns are difficult to recognize and allow us to distinguish these pattern individual faces from several angles. Processing of stored into as patterns and then utilize this pattern for problem solving is a new field in computing[8]. Artificial neural networks are a group of models inspired by biological neural networks and used to guess functions[9] which depends upon a large number of unknown inputs. In artificial neural networks, there are systems of interconnected "neurons" which exchange messages between collectively. Zeng *et al.*[11] predicted the superconducting and non-superconductors transition temperatures for different compounds through data-enhanced technology by developing convolutional neural networks. Ali *et al.*[12] predicted the comparison of photovoltaic nano fluid and nano-PCM system using artificial neural network. Shiet *et al.*[13] predicted the mechanical and electrical properties of engineered cementitious composite for the design of efficient material. They also discussed the parameters which affects the performance of artificial neural network. Ahmadi *et al.*[14] used two artificial neural network algorithms to predict relative thermal conductivity of Al2O3/Water nanofluid and compared the effects of the temperature, concentration and particle size. Qi *et al.*[15] used ANN and particle swarm optimization methods to predict the unconfined compressive strength of cemented paste backfill strength. Varol *et al.*[16] used ANN model to predict the degrees of accuracy for density and hardness of AA2024-SiC nanocomposites. Shahsavar *et al.*[17] applied group method of data handling neural network to develop the correlations for liquid paraffin-Fe_3O_4 thermal conductivity and viscosity. Pal *et al.*[18] modified Lewis-Nelsen model, according to this model electrical conductivity of plastic considering electrically-conductive filler particles (carbon) into plastic matrix and thermal conductivity of plastic can be enhanced. The mathematical relation is given by:

$$K = K_c\left[1-\frac{\phi}{\phi_m}\right]^{-3\phi_m} \quad \ldots (1)$$

where ϕ_m is the maximum packing concentration, K-effective electrical conductivity and K_c is conductivity of composite.

Cai *et al.*[19] used percolation model developed to describe the effective electrical conductivity of particle filled composites with the help of effective medium and given by below percolation equation:

$$\sigma_{eff} = \sigma_1\left(\frac{\phi-\phi_m}{\phi_m}\right)^{-s}, \phi > \phi_m \quad \ldots (2)$$

where, σ_{eff} is the effective conductivity of composite, ϕ_m is maximum packing concentration } and *s* is percolation threshold. σ_1 is conductivity of insulator. Zhang *et al.*[20] used Monte-Carlo simulations for the prediction EEC of short fiber composites and demonstrate the relationships between the conductivity and different dependent parameters. Torquato *et al.*[21] investigated that effective conductivity in any dispersion is higher provided that the average size of the cluster in dispersed phase is far smaller than the size of the sample. Miller *et al*.[22] studied the effective properties (magnetic, electrical, thermal) for statistically isotropic and homogeneous materials. The cumulant method and the effective medium theory are two numerical methods developed by Hori *et al.*[23] to predict the EEC. Their findings are matched with the data of computer simulation to compute the effective conductivity of a three dimensional random network. Hori *et al.*[24] also studied that perturbed solution can be acquired by many-point correlation functions of the permittivity field. Hori *et al*. also applied perturbed extensions to predict the effective permittivity with the help of electric displacement, electric field, Lorentz field and *T* matrix and debate several approximate solutions involving the actual-medium estimate[25]. Hori *et al.* predicted the effective permittivity of random heterogeneous materials and expressed it in terms of many-point correlation functions of the spatial variation of permittivity[26]. Río *et al*.[27] investigated the porous Silicon model in which EEC of crystalline Si rises to a maximum value at an optimum porosity. Jin *et al*.[28] performed simulation for the study of phase field model to predict EEC in complex microstructures. Weber *et al.*[29] experimentally and analytically determined the electrical properties of Al-Si eutectic alloy. Gabaldón *et al.*[30] studied the effect of porosity on the EEC of the ceramic membranes which increases with the decrease of pressure. Brederlow *et al.*[31] studied, that EEC can be reduced by streamers, as the gas pressure is increased. Zamel *et al*.[32] studied the effect of the porous assembly on the EEC of three-dimensional carbon paper of long cylindrical fibers. In the past few years, many theoretical models[33-37] are investigated to estimate the EEC of composites. Glorieux *et al.*[38] employed the neural network approach to study the eddy-current inverse problem. Tsai *et al.*[39] studied the influences of conversion of polarity on the electrode of the electrical discharge machining process. Therdthai *et al.*[40] established a non-linear relationship with the help of ANN approach and studied the effect of milk ingredients and temperature on the electrical conductivity. Hezave *et al.*[41] investigated the applicability of artificial neural network to predict the electrical conductivities of ternary systems comprising with PF6, water and ethanol. Sarkar *et al.*[42] used ANN approach to study the wire electrical discharge machining of gamma titanium aluminide that depends upon surface roughness and cutting speed. Artificial neural network approach is also used to predict the EEC of complex composites used for different applications[43,44]. Graphite-filled wax/polyethylene blends materials are synthesized for the thermal energy storage applications[45,46].

In the present research, artificial neural network is well appropriate for the complex behavior of particulate polymer composites[5,6]. There are limited experimental and theoretical evaluations on the EEC polymer composites filled by fiber that would demand exact value of the EEC of polymer composites. This work employee an ANN for the prediction of EEC of copper powder filled low density polyethylene and linear low density polyethylene composites.

2 Back Propagation Algorithm (FFBP)

ANNs can be classified into feedback and feedforward networks. In feedback networks, connections between processing elements are in both the forward and backward directions. In feedforward networks, the connections between the processing elements are in the forward direction only. The feedforward network also called multilayer perceptron and is trained with the backpropagation algorithm, and radial basis function. A multilayer feedforward network learns by backpropagation, in which error propagates back is called feedforward backpropagation (FFBP).

The following mathematical equations are used to model the FEBP algorithm:

(i) The difference between actual and desired activity is called derivative error (DE) which is given by:

$$DE_k = \frac{\partial E}{\partial y_k} = y_k - d_k \quad \dots (3)$$

(ii) When step first is the multiply by of the rate of change of output per unit as total change in input occurs that gives quantity (Q) which is given by:

$$Q_k = \frac{\partial E}{\partial X_k} = \frac{\partial E}{\partial y_k} \times \frac{\partial y_k}{\partial x_k} = DE_k y_k (1 - y_k) \quad \dots (4)$$

(iii) When the step second is multiply by activity level of unit that gives quantity (S) which is given by:

$$S_{jk} = \frac{\partial E}{\partial S_{jk}} = \frac{\partial E}{\partial X_k} \times \frac{\partial X_k}{\partial S_{jk}} = Q_k y_j \quad \ldots (5)$$

(iv) When weight on connection is multiply by the step third that gives output unit which is given by:

$$A_j = \frac{\partial E}{\partial y_j} = \sum_k \frac{\partial E}{\partial x_k} \times \frac{\partial x_k}{\partial y_j} = \sum_j Q_k S_{jk} \quad \ldots (6)$$

Two activation functions are used for **ANN approach**

(A) A Pure Linear function (PURELIN) is given by:

$$Y_k = AX_k \quad \ldots (7)$$

(B) A Tangent Sigmoid function (TANSIG) which can be written as:

$$Y_k = \frac{2}{(1+\exp(-2X_k)) - 1} \quad \ldots (8)$$

The activity of output layer is given by two steps:

First, it gives the total weight by the formula

$$X_k = \sum_j y_j W_{jk} \quad \ldots (9)$$

where, y_j is the activity of k^{th} level and W_{jk} is the weight between j^{th} and k^{th} unit. Second, it finds out the activity Y_k by employing some mathematical functions.

3 Training Algorithms

There are different backpropagation training algorithms in MATLAB. The details of the training functions are described in Table 1.

4 Results and Discussion

The LDPE and LLDPE based composites filled with copper have a complex inner structure. Therefore, it may not be practical to describe all details of the structure accurately. The effective electrical conductivity depends on various characteristics of the material, accounting for all these in order to predict effective electrical conductivity is a tedious task either numerically or theoretically, and even more difficult to establish a real model for the non-linear problem. The complex geometry along with the large difference in the electrical conductivity of the constituents makes it difficult to calculate the effective electrical conductivity. For this reason, artificial neural networks have been utilized in the case of copper powder filled low density polyethylene.

The study of nonlinear behaviour of EEC is shown by using ANN approach. The six different training functions of ANN approach are used to study of nonlinear behavior of the EEC. The mapping of input and output pattern is done by the help of FFBP networks.

A three layer feed forward network is used for prediction of the EEC of LDPE and LLDPE based composites filled with copper. The network's input has range from one to three. The first layer contains two TANSIG neurons, whereas second one contain one PURELIN, 1000 epochs are run in TANSIG-PURELIN threshold function. The final layer is third layer that is output layer for six training functions of the FFBP network. The ANNs give very excellent result of effective electrical conductivity of the copper filled LDPE and LLDPE composites

5 EEC of LLDPE/Cu

The experimental values of EEC[37] of LLDPE/Cu composites are plotted by using different training functions (TRAINOSS, TRAINLM, TRAINBR, TRAINSCG, TRAINBFG and TRAINRP) of ANNs approach. Figures 2-7 show the variation in the experimental value of EEC LLDPE/Cu those predicted with the help of the training function of ANN and other models with the volume fraction of filler (copper). Over a wide range (0 to 24%) of

Table 1 — Explanation of the different training algorithms.

Training	Function	Description
TRAINSCG	Scaled conjugate gradient backpropagation	general-purpose training algorithm
TRAINOSS	One-step secant backpropagation	Compromise between conjugate gradient methods and quasi-Newton methods
TRAINLM	Levenberg–Marquardt backpropagation	It is the fastest training algorithm for networks of moderate size.
TRAINBR	Bayesian regularization	Modification of the Levenberg–Marquardt training algorithm
TRAINBFG	BFGS quasi-Newton backpropagation	It has more computation in each iteration than conjugate gradient algorithms
TRAINRP	Resilient backpropagation	It updates weight and bias values according to the resilient backpropagation algorithm

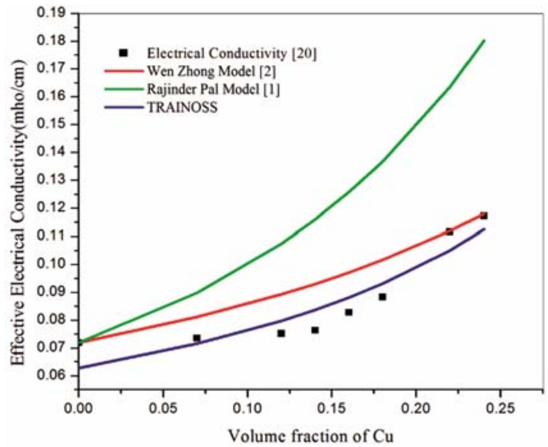

Fig. 2 — EEC of LLDPE/Cu with training function TRAINOSS.

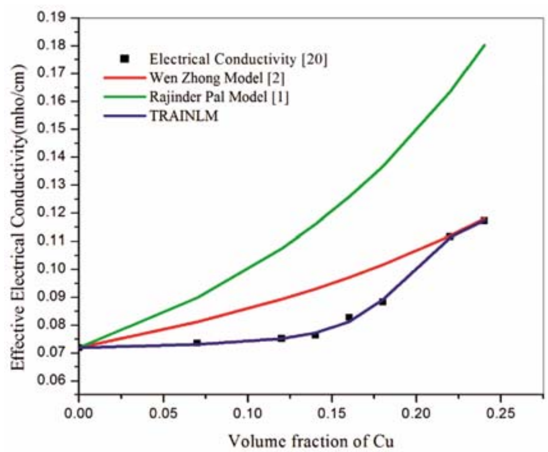

Fig. 3 — EEC of LLDPE/Cu with training function TRAINLM.

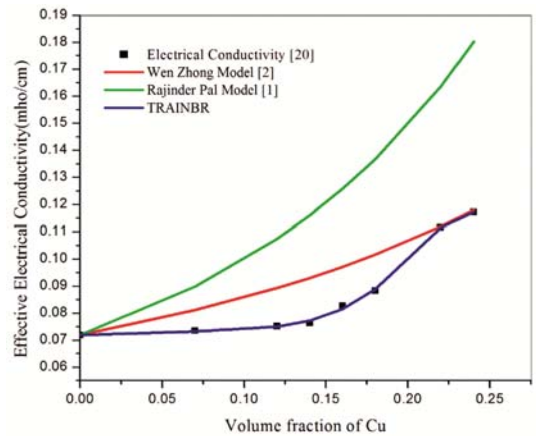

Fig. 4 — EEC of LLDPE/Cu with training function TRAINBR.

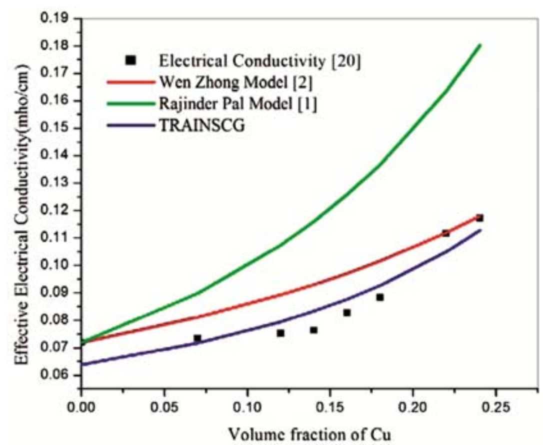

Fig. 5 — EEC of LLDPE/Cu with training function TRAINSCG.

volume fraction of copper (filler) are taken in the Figs 2 to 7. All training function of artificial neural network gives the one output that is EEC. It is observed that the EEC of composites increases directly with filler loading. The maximum value of EEC is 0.1204 mho/cm (TRAINBFG) which is achieved for LLDPE containing 24% volume fraction of copper. The increment in the EEC of LLDPE/Cu composites was expected, as EEC of filler (Cu) was significantly higher ($\sigma_{Cu=}$ 15.89 mho/cm) than that of LLDPE (σ_{LLDPE}=0.1204 mho/cm). With the increase in volume content of copper in LLDPE, Cu particles improved and start interacting with each other, which results in the increase in EEC by increasing the volume fraction of copper. Many models failed to predict the EEC of LLDPE/Cu composites over a full range of filler concentration used. From all the graphs it can be observed that the calculation of R. Pal model (ϕ_m=0.6) does not give the satisfactory result of EEC[18]. While Wen Zhong model (ϕ_m=0.6 and s=0.97) gives satisfactory result with experimental and predicted EEC at the higher concentration of filler (22% and 24%)[19]. Figure 8 shows that the percentage deviation in EEC using different functions of ANN for the LLDPE/Cu. TRAINBR and TRAINLM give least deviation among the all functions. TRAINOSS and TRAINSCG provide the maximum deviation between all the training functions.

6 EEC of LDPE/Cu

The ANN approach which contains six different functions (TRAINOSS, TRAINLM, TRAINBR, TRAINSCG, TRAINBFG, and TRAINRP) has been employed and the EEC of LDPE/Cu is plotted. Figures 9-14 show the changes in the experimental data[37] of EEC LLDPE/Cu those predicted by the training function of ANN and other models with the volume fraction of filler (copper). The volume fraction of copper (filler) 0 to 24% is taken over a wide range in Figs 9-14. Six different training functions of ANN gives the one

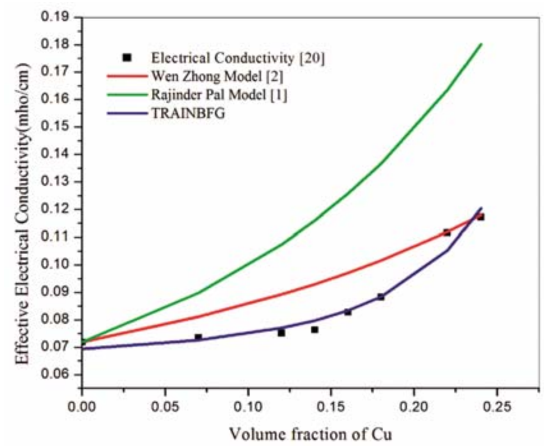

Fig. 6 — EEC of LLDPE/Cu with training function TRAINBFG.

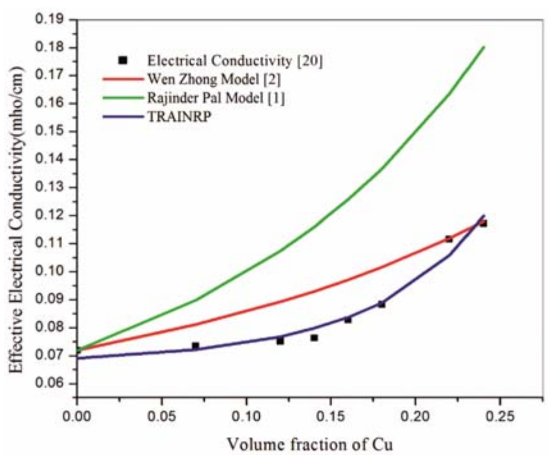

Fig. 7 — EEC of LLDPE/Cu with training function TRAINRP.

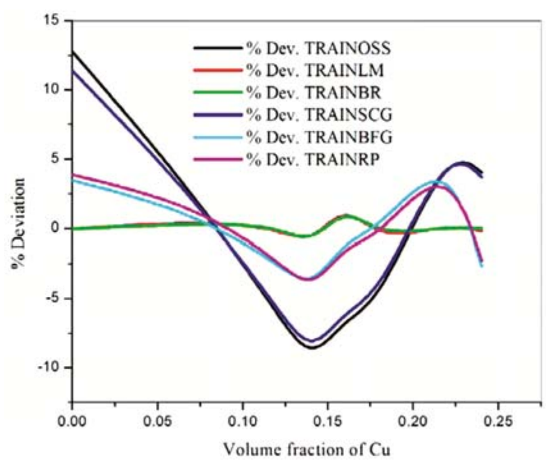

Fig. 8 — Deviation (%) in EEC using different training functions of ANN for the LLDPE/Cu.

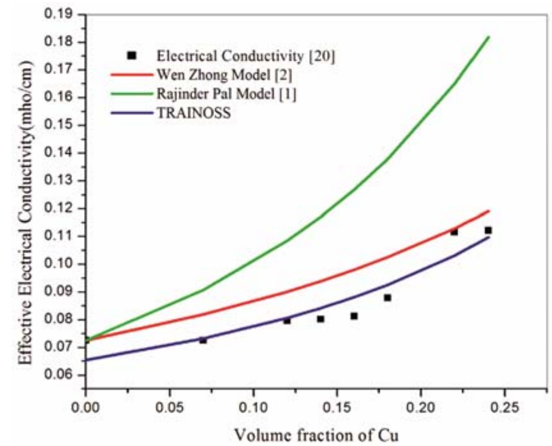

Fig. 9 — EEC of LDPE/Cu with training function TRAINOSS.

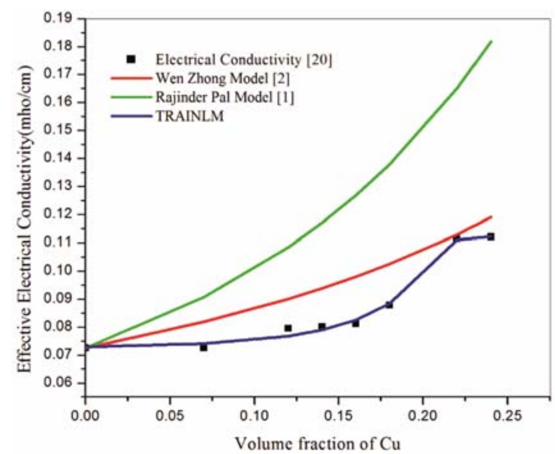

Fig. 10 — EEC of LDPE/Cu with training function TRAINLM.

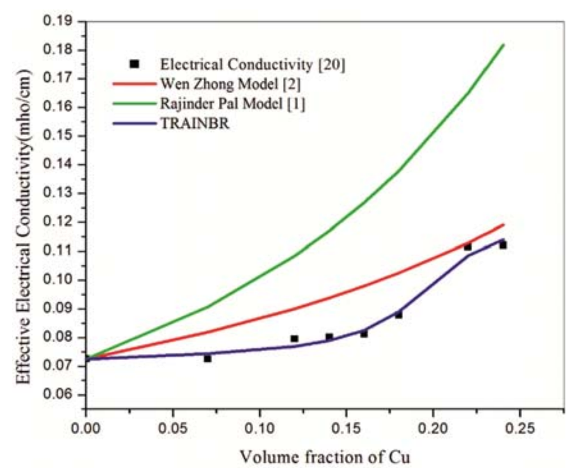

Fig. 11 — EEC of LDPE/Cu with training function TRAINBR.

output. It seems that the EEC of the composites increases with the increment in filler loading. The maximum value of EEC is 0.1160 mho/cm (TRAINBFG and TRAINRP) which is achieved by for LDPE having 24% volume fraction of copper. As the volume of filler increases the content of LDPE also increases because the molecule of filler interacts with each other resulting in an increase in EEC as

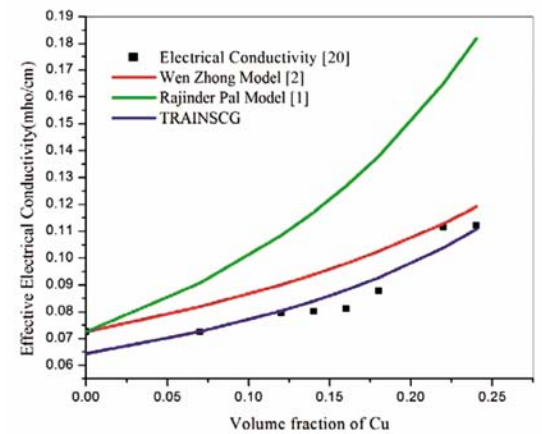

Fig. 12 — EEC of LDPE/Cu with training function TRAINSCG.

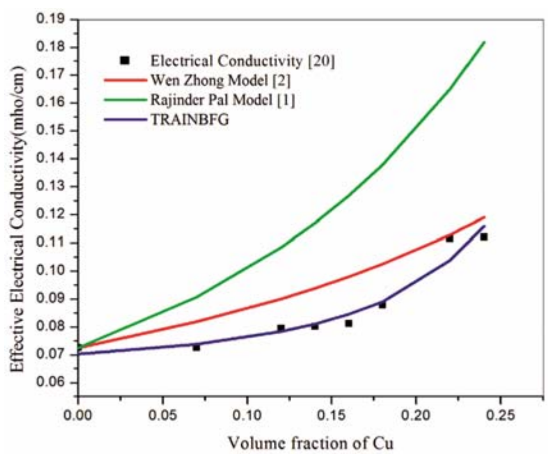

Fig. 13 — EEC of LDPE/Cu with training function TRAINBFG.

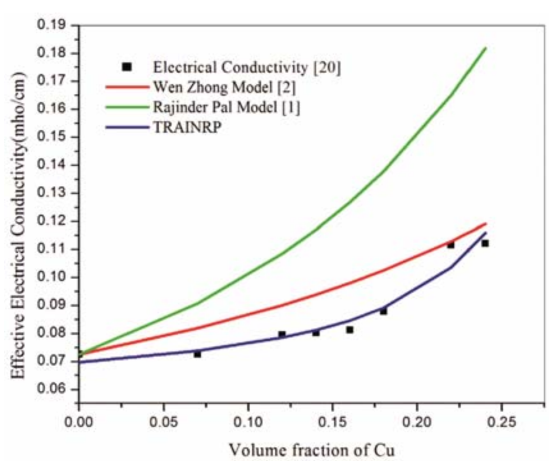

Fig. 14 — EEC of LDPE/Cu with training function TRAINRP.

volume fraction of copper increases. The increment in the EEC of LDPE/Cu composites was expected, as the EEC of LLDPE (σ_{LLDPE}=0.1160 mho/cm) is lower than filler (Cu) ($\sigma_{Cu=}$ 15.89 mho/cm. Many techniques failed to predict the EEC of LLDPE/Cu

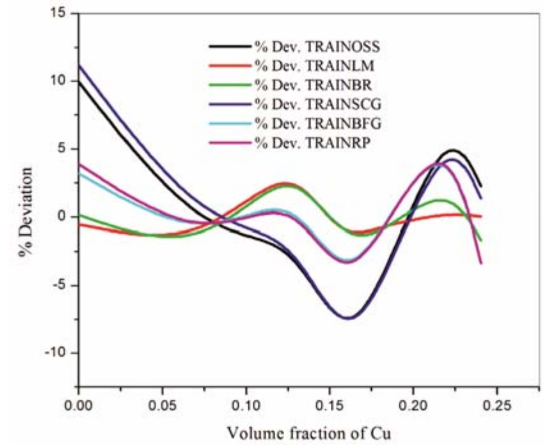

Fig. 15 — Deviation (%) in EEC using different training functions of ANN for the LDPE/Cu.

composites over a full range of filler concentration used. From all the graphs, we observed that the calculation of R Pal model[18] (ϕ_m=0.6) does not give the satisfactory result with EEC. While the calculation result by Wen Zhong model[19] (ϕ_m=0.6 and s=0.97) of equation is satisfactory in agreement with experimental and predicted EEC at the higher concentration of filler (at 22%). At the very high concentration our predicted result does not match with Wen Zhong model. Prediction of EEC by using different training functions of ANNs gives satisfactory results. Figure 15 shows the percentage deviation in EEC using different functions of ANN for the LDPE/Cu. TRAINOSS and TRAINSCG give the most deviation among all the training functions. TRAINBR and TRAINLM give least deviation among the all functions.

7 Conclusions

A 3 layered feed forward neural network that is fully connected with the succeeding layer through connection weights is used for the prediction of EEC of copper filled with LDPE and LLDPE composites. It has been shown that the ANN method has a good prediction capability for nonlinear behaviour of EEC of copper filled LDPE/LLDPE composites. In comparison with other unfilled polymer, EEC increases in filler content of polymer. The EEC of particulate polymer composites is dependent on input parameters. In this work, we have used six training functions (TRAINOSS, TRAINLM, TRAINBR, TRAINSCG, TRAINBFG and TRAINRP) of ANN network. The reported result on EEC of copper powder filled LDPE/LLDPE composites by different training functions of ANN approach agreed perfectly with experimental value. In comparison with existing theoretical models, the present ANN technique does not demand any additional empirical parameter. Hence, it has good modelling efficiency for a new 3 or more phase complex materials. It is well-known that the ANN approach can be comprehensive to investigation of further material's EEC of particle filled composites.

Acknowledgment

One of the author Sukhmander Singh appreciates the financial support given by the University Grants Commission (UGC), New Delhi, for providing the startup Grant (Project No. F. 30-356/2017/BSR).

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