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RESEARCH ARTICLE

Development of Series-Parallel and Neural-Network Based Models for Predicting Electrical Conductivity of Polymer Nanocomposite

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ABSTRACT Polymer nanocomposites are emerging hybrid materials for the production of energy storage electrodes, biomedical sensors, and building construction materials. However, experimentation cost and time can be unfavorable to their performance investigation. Therefore, using a modeling approach to predict the electrical conductivity of polymer nanocomposite is an effective approach in mitigating experimentation cost and time. Since the polymer nanocomposites' electrical conductivity depends on several factors, the engagement of efficient analytical models for predicting their properties, cannot be overemphasized. Herein, this study developed a series-parallel model, which incorporates the connection between the polymer and the nanofillers for the prediction of the electrical conductivity of graphene-polypyrrole (Gr-PPy) and reduced graphene oxide/polyvinyl alcohol/polypyrrole (RGO/PVA/PPy) nanocomposites. In addition to explicit modelling, an artificial intelligence approach (neural network) was also explored for the prediction tasks. The results of the models in an entity and when compared to an existing model, show flexibility and accuracy for the polymer nanocomposites electrical conductivity prediction. It can be inferred that the model can be suitable to predict the electrical conductivity of polymer nanocomposites.

INDEX TERMS Electrical conductivity, energy storage, graphene, modeling, nanocomposite, polymer.

I. INTRODUCTION

Graphene (Gr) and its various derivatives are suitable filler materials for conductive polymers to achieve high electrical conductivity for battery electrodes. As a carbonaceous material, graphene and graphene derivative polymer nanocomposites are characterized by exceptional electronic properties due to their high electrical conductivity [1] and porosity [2]. Besides the electrical conductivity enhancement of polymers, graphene and its derivatives offer a wide range of applications due to their flexibility and exceptional mechanical, thermal, and chemical properties [3].

Polypyrrole (PPy), a conducting polymer, is a multifunctional polymer whose applications extend to the manufacturing of electronic devices, such as optoelectronic devices [4], sensors, water waste remover [5], and supercapacitors [6]. However, polypyrrole has a poor cycle life in charge/discharge duty and poor thermal stability at high temperatures [7]. For this reason, the electrochemical and thermal properties of polypyrrole must be tuned as a means to

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effectively exude the full advantage of the valuable properties of the polymer.

Polyvinyl alcohol (PVA) on the other hand, is a biocompatible/biodegradable polymer, possessing quality properties, such as: flexibility, nontoxicity, high tensile strength and thermal/chemical stabilities [8]. In addition, PVA has the advantage of solubility in water: the solubility property of PVA makes it dispersible on graphene, metal oxides, and polymers to form homogenous composites. Nevertheless, PVA is a nonconducting polymer, a scenario that hinders its utilization in many electronic devices. However, the excellent mechanical, thermal, and chemical properties are beneficial to electrochemical electrodes when composited with suitable fillers. In this case, the intrinsic properties of the polymer is retained while synergistically functioning in polymer nanocomposite electrodes [9].

The properties of polymer nanocomposites act as prominent factors responsible for their versatile application areas. This, is due to the possibility of tuning the properties of polymers by introducing polymers, metal/metal oxides, and carbonaceous materials. For energy storage electrodes, the electrical conductivity of electrodes must be high enough in order to accommodate the high energy demand of electric vehicles and grid energy load balancing. A very high current rate, which frequently occurs when a large number of charge carriers are needed, is a function of the electrical conductivity of the electrode active materials. Hence, the electrochemical capacity and performance of batteries depend on the electrical conductivity of the electrodes: a low electrical conductivity suggests low-capacity electrodes and vice versa [10]. Therefore, studying the electrical conductivity of polymer nanocomposite electrodes is essential to their performance specifications. The total resistance of polymer nanocomposite can be represented as the sum of the tunneling and intrinsic resistance, as presented in Equation (1), [11], [12].

$$R_{pc} = R_i + R_t \tag{1}$$

where R_i, R_t, and R_{pc}, are polymer nanocomposite electrodes' intrinsic, tunneling, and effective resistance. Generally, the electrical conductivity of polymer nanocomposite is controlled by filler volume fraction, size, shape, orientation, porosity, and aspect ratio [13], [14]. These factors can be thoroughly understood by modeling and computer simulations. Modeling and computer simulation of the electrical conductivity of polymer nanocomposites will reduce experimentation time, cost, and susceptible errors.

This study modeled and simulated the electrical conductivity of graphene/polypyrrole (Gr-PPy) and reduced graphene oxide/polyvinyl alcohol/polypyrrole (RGO/PVA/PPy) nanocomposites. The electrically conductive network of the nanocomposite is subject to several factors, such as: the intrinsic properties of the filler and the polymer matrix, and the preparation method. The filler aspect ratio is another factor which influences the effective properties of composite materials [15]. Percolation theory is often used to explain that at any region of the composite, there is a likelihood for conduction to take place due to the sufficient interconnect of the particles [16]. The percolation path formed by the random distribution of conductive filler on the polymer matrix, is most suitable for proper characterization of the composite. When the percolation threshold is attained at low quantity of conductive filler, the composite can be said to be kinetically percolated [17]. The mathematical modeling of the electrical property of polymer nanocomposite, is the most viable means to observe the impact of the various factors on which the property polymer nanocomposites rely. The percolation threshold and the saturation point of the polymer nanocomposite can be adequately determined using modeling. Therefore, to further explore the versatility of models in characterizing the electrical properties of polymer nanocomposites, two modeling approaches were developed and engaged in this study, to characterize the electrical conductivity of polypyrrole, polyvinyl alcohol, and graphene nanocomposites. The first modeling method is an analytical approach which considers the random series and parallel connections of the nanoparticles. The second approach is the use of artificial neural network. The artificial neural network is expected to precisely present and predict the dynamic relationship of polymer nanocomposites input and output [18].

Furthermore, the aim of this study is to propose the series-parallel model as well as an artificial neural network approach for the prediction of the electrical conductivity of polymer nanocomposite. The objectives include: (1) the development of a novel electrical conductivity model based on the series and parallel contact of polymer and additives (2) the quantification and verification of the developed model by using a machine learning approach. The model formulation is presented in Section II; Section III provides the materials and experimentation of the investigated polymer nanocomposites. Section IV gives the details results and discussion of the study. Concisely presented in Section V is the conclusion of the study. The results of the models showed good correlation with experimental measurements.

II. MODEL FORMULATION

Based on the industrial applications of polymer nanocomposites, ample experimentations have been conducted on the investigations of their various properties, such as electrical conductivity, mechanical strength, and thermal and chemical stabilities [19], [20]. However, experimentation alone cannot provide enough information about the nature of polymer nanocomposites. Theoretical models are useful tools in quantitatively predicting the effective electrical conductivity of polymer nanocomposite. In addition, the parametric analysis of the composite can be intensively, precisely, and concisely carried out with high accuracy.

Mixture models have been applied in earth science to analyze the electrical conductivity properties of soil. For example, Archie's mixture model has been widely engaged to observe the relationship between soil electrical conductivity and water content [21]. The modified Archie's law of mixing model, which can predict the electrical conductivity of the two-conducting phase, is presented in Equation (2) [22].

$$\sigma = \sigma_1(1-\lambda)^p + \sigma_2\lambda^m \tag{2}$$

where σ_1, σ_2 are the conductivities of the insulating and the conducting phase; p and m are connecting exponents; λ is material's volume fraction.

A. SOME EXISTING MODELS FOR POLYMER COMPOSITES ELECTRICAL CONDUCTIVITY PREDICTION

In previous years, several models have been developed to characterize the electrical conductivity of polymer composites. The power law model, shown in Equation (3), is one of the earliest models for predicting polymer composites' electrical conductivity [23].

$$\sigma = \sigma_0(V - V_p)^t \tag{3}$$

where σ, σ_0, V, and V_p, are the composite electrical conductivity, filler intrinsic electrical conductivity, filler volume fraction, and percolation threshold; t is the connecting exponent. The power law model presents an established basis for the prediction of the insulator-semiconductor-conductor transition of polymer and their nanocomposite. However, the power law model is not precisely accurate for the prediction of polymer composite electrical conductivity [24]. In a broader terms, the contact resistance between conductive filler and polymer matrix determines the effective electrical conductivity of the polymer composite. Therefore, Kovacs et al. [25] introduced resistances within the transitive conduction regions of polymer composite. As shown in Equation (4), the model of Kovacs et al., was set-up to contain the radius (r) and resistance (R) of the particles and the contact resistance (Rc) [25].

$$\sigma = \frac{V\phi^{(2x+1)}}{R + Rc};\, V = \frac{l}{2\pi r^2} \tag{4}$$

where ϕ is the filler weight fraction, and x is the control exponent. One of the deficiencies of this model is its lack of consideration of the matrix's intrinsic parameters. Moreover, the electron tunneling between a filler and matrix, is a function of the contact between the nanoparticles. In other words, the transition of a polymer from an insulator to conductor or from semiconductor to conductor, largely depends on the contact between the composite nanoparticles.

As presented in Equation (5), Deng and Zheng [26] developed an analytical model which considered random orientation of carbon-nanotube (f), filler and matrix conductivities (σ_f, σ_p), and effect of contact resistance (μ^2). According to Arjmandi et al. [27], Deng and Zheng model is insufficient to describe the effects of tunneling and contacts in polymer composite electrical conductivity.

$$\sigma = \frac{\mu^2 f \sigma_f}{3} + \sigma_p \tag{5}$$

B. THE PROPOSED SERIES-PARALLEL MODEL

The conductivity of polymer nanocomposites can be treated in a similar manner to the conductivity of soil in relation to water content. However, the complex distribution of fillers on a polymer matrix requires a simple but precise model for the proper characterization of polymer nanocomposite properties. The properties of polymer nanocomposites are functions of the properties of the materials composited [28]. Two mixture scenarios can be deduced from the contact of the composites, which are:

1) series contact
2) parallel contact

First, the model of Kirkpatrick [29] was considered to derive the series-parallel model proposed in this study. Kirkpatrick stated that the random arrangement of composite materials, influences their electrical conductivities in diverse ways. The random arrangement can either be in series or parallel, or both. The conducting filler and the polymer matrix with a weight fraction of ϕ, is expected to have a conducting weight fraction of $1-\phi$. Rhodes et al. [30] argued that bulk composite (e.g. soil) electrical conductivity results from parallel conductors of liquid-phase and bulk conductivities of the materials. That is, the electrical conductivity of the composite is a function of the tunneling of electrons within the pores of the composite materials, the intrinsic conductivities of the materials, and the interfacial resistances. The electrical conductivity of polymer nanocomposites, can be treated in the light of Rhodes et al. argument. Hence, in this study, polymer nanocomposite electrical conductivity is assumed to be as a result of the contribution of series and parallel random contacts of composited materials.

Therefore, for an effective prediction, the electrical conductivity of the composite can be estimated by the addition of the series and parallel contact of the materials. For the parallel connection of the materials, Equations (6)&(7) are as presented. The formulated electrical conductivity equation follows the assumption of literature [31].

$$\frac{1}{\sigma_c} = \left(\frac{1}{\sigma_f}\right)\phi_1 + \left(\frac{1}{\sigma_p}\right)\phi_2 \tag{6}$$

$$\frac{1}{\sigma_c} = \frac{\sigma_p\phi_1 + \sigma_f\phi_2}{\sigma_f\sigma_p} \tag{7}$$

where σ_c, σ_f and σ_p are the electrical conductivity due to parallel contact, filler electrical conductivity, and polymer electrical conductivity; ϕ_1 and ϕ_2 are the filler and polymer weight ratios.

The series connection assumes that the electrical conductivity, σ_s, is a sum of the contributing materials with respect to their volume fraction, as given in Equation (8) [32].

$$\sigma_s = \sigma_f\phi_1 + \sigma_p\phi_2 \tag{8}$$

Therefore, the effective electrical conductivity can be written as presented in Equation (9):

$$\sigma_{eff} = \sigma_c + \sigma_s \tag{9}$$

hence,

$$\sigma_{eff} = \left(\frac{\sigma_p\phi_1 + \sigma_f\phi_2}{\sigma_f\sigma_p}\right)^{-1} + \sigma_f\phi_1 + \sigma_p\phi_2 \tag{10}$$

$$= \frac{\sigma_f\sigma_p + (\sigma_f\phi_1 + \sigma_p\phi_2)(\sigma_p\phi_1 + \sigma_f\phi_2)}{\sigma_p\phi_1 + \sigma_f\phi_2} \tag{11}$$

$$\sigma_{eff} = \frac{\sigma_f\sigma_p(1 + \phi_1^2 + \phi_2^2) + \phi_1\phi_2(\sigma_f^2 + \sigma_p^2)}{\sigma_p\phi_1 + \sigma_f\phi_2} \tag{12}$$

Dividing the numerator and denominator of Equation (12) by $\sigma_p\phi_1$:

$$\sigma_{eff} = \frac{\sigma_f(1 + \phi_1^2 + \phi_2^2)\phi_1^{-1} + (\sigma_p^2 + \sigma_f^2)\phi_2\sigma_p^{-1}}{1 + \frac{\sigma_f}{\sigma_p}\left(\frac{\phi_2}{\phi_1}\right)} \tag{13}$$

And substituting the non-variable values as: ρ, μ, and δ:

$$\sigma_{eff} = \frac{\rho(1 + \phi_1^2 + \phi_2^2)\phi_1^{-1} + \mu\phi_2}{1 + \delta\left(\frac{\phi_2}{\phi_1}\right)} \tag{14}$$

The mixing condition obeys [33] and it is as shown in Equation 12.:

$$\phi_1 + \phi_2 = 1; \therefore \phi_2 = 1 - \phi_1; 0 < \phi_1 < 1; 0 < \phi_2 < 1 \tag{15}$$

Therefore, Equation (16) gives the effective electrical conductivity of the polymer nanocomposite without connecting exponent.

$$\sigma_{eff} = \frac{2\rho(1 - \phi_1 + \phi_1^2)\phi_1^{-1} + \mu(1 - \phi_1)}{1 + \delta\left(\frac{1-\phi_1}{\phi_1}\right)} \tag{16}$$

By applying the connecting exponent, t [29], Equation 16 becomes:

$$\sigma_{eff} = \frac{2\rho\left((1 - \phi_1 + \phi_1^2)\phi_1^{-1}\right)^t + \mu(1 - \phi_1)^t}{1 + \delta\left(\frac{1-\phi_1}{\phi_1}\right)^t} \tag{17}$$

The series-parallel polymer nanocomposite electrical conductivity model, derived in Equation (17), is expected to link all the conducting channels in the nanocomposite and accurately predict the saturation point. In this case, the weight fraction (ϕ) is the independent variable, while other parameters, are calculated.

C. PROPOSED ARTIFICIAL NEURAL NETWORK (ANN) ARCHITECTURE

Artificial neural networks are computational tools that mimic human brain behavior by storing experiential knowledge and being able to reproduce it for use at any time [34]. An artificial neural network is a cognitive model; therefore, it can potentially model the nonlinear relationship between the electrical conductivity of polymer composites and the influential parameters. The electrical conductivity of polymer nanocomposite is usually a nonlinear curve with respect to volume fraction. Hence, the applicability of artificial neural networks to polymer composites' electrical conductivity prediction is significant. The artificial neural network model's general benefits include: cognitive ability relating to precise prediction; effective nonlinear statistical modeling; and its ability to manipulate large data sets [35], etc. In this study, the independent parameters considered in the artificial neural network model are the calculated values of the parameters from the series-parallel model. Figure 1 shows the study artificial neural network model.

The study model, shown in Figure 1, was trained by the Levenberg-Marquardt algorithm (LMA). As shown in

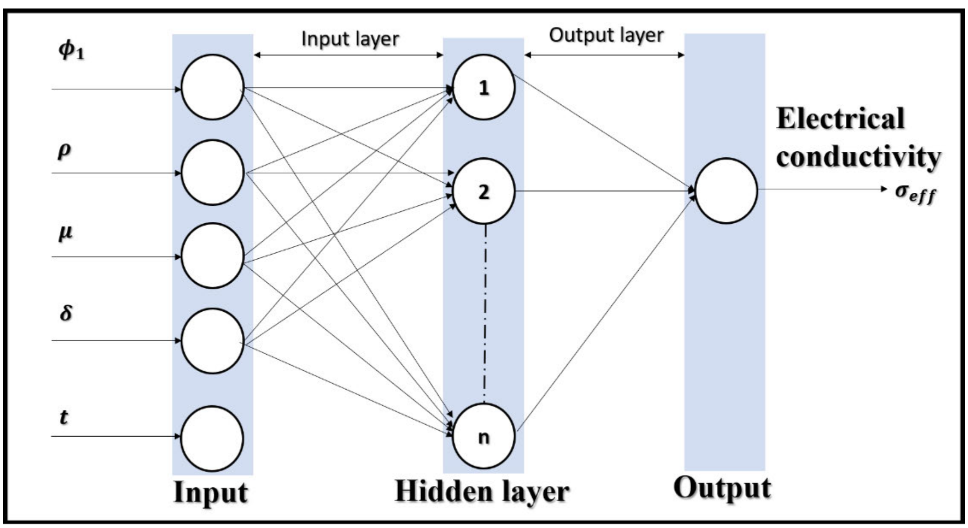

FIGURE 1. Study artificial neural network model.

Figure 1, the input, hidden, and output processes are allocated with input neurons, a transfer function, and output neurons. Levenberg-Marquardt is an algorithm developed to solve nonlinear regression problems; the model is a combination of Gauss-Newton and gradient descent methods [36]. In this study, the Levenberg-Marquardt algorithm, shown in Equation (18), was used to train the tangent sigmoid transfer function, Equation 19 [35], [37].

$$y_i = \frac{1}{1+e^{-x_i}} \tag{18}$$

$$\left(J^T J + \lambda I\right) h_l m = J^T E \tag{19}$$

where y_i and x_i, are the output of the neuron and the weighted sum of input to neuron; J is the Jacobian matrix, λ is the dampen-parameter, I is the identity matrix, E is the error vector, and h_{lm} is the input parameter adjuster.

III. MATERIALS AND EXPERIMENTATION

The datasets for the series-parallel electrical conductivity model were obtained from laboratory experimentation of graphene nanoplatelets loaded-polypyrrole [38] and reduced graphene-oxide loaded polyvinyl alcohol/polypyrrole nanocomposites [39].

The preparation method of the nanocomposites involves the use of solvent solution blending. The exfoliated graphene in the composite of Gr-PPy, was dispersed in deionized-water by ultrasonication process; furthermore, the mixture of the exfoliated graphene and polypyrrole, was magnetically stirred, filtered, and dried. In addition, the production of RGO/PVA/PPy nanocomposite, was achieved by the dissolvement of PVA in deionize water under magnetic stirring. The mixture of PPy and RGO obtained by ultrasonication process, was loaded onto the PVA, magnetically stirred, filtered, and vacuum-dried. The measured electrical conductivity of the samples, are as shown in Figures 2&3. Note that the electrical conductivity of Gr-PPy nanocomposite data, was smoothened by 3-order polynomial function. The processed electrical conductivity data for RGO/PVA/PPy nanocomposite, has twenty-one-data points, while Gr-PPy nanocomposite, has seven-data points.

IV. RESULTS AND DISCUSSION

The electrical conductivity of polymer nanocomposite models, are usually used to describe the non-linear behavior and the dependency of the electrical conductivity of polymer nanocomposite on the volume fraction of nanocomposite filler. In addition, the effect of electrical conductivity on energy storage is directly proportional to the performance rate of the storage electrode.

As shown in Figure 2, the electrical conductivity measurement of the Gr-PPy nanocomposite displayed a percolation threshold at about 0.15 wt (%) of the graphene content in the conductive polymer matrix. The linear characteristics of the conductivity with respect to the graphene weight fraction begins at about 0.15 wt (%) to 0.25 wt (%). The

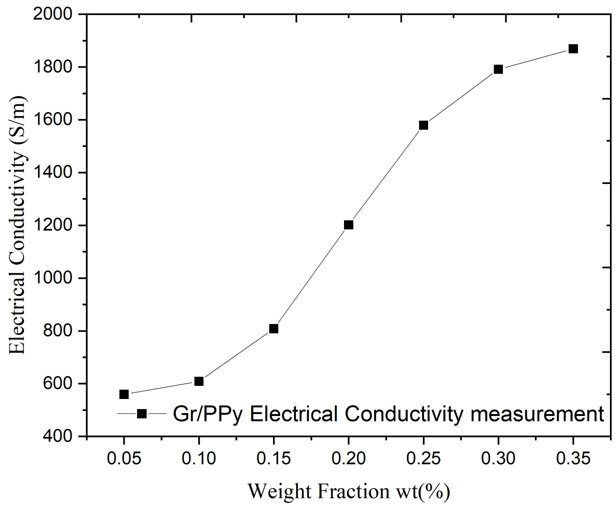

FIGURE 2. Gr-PPy nanocomposite electrical conductivity data.

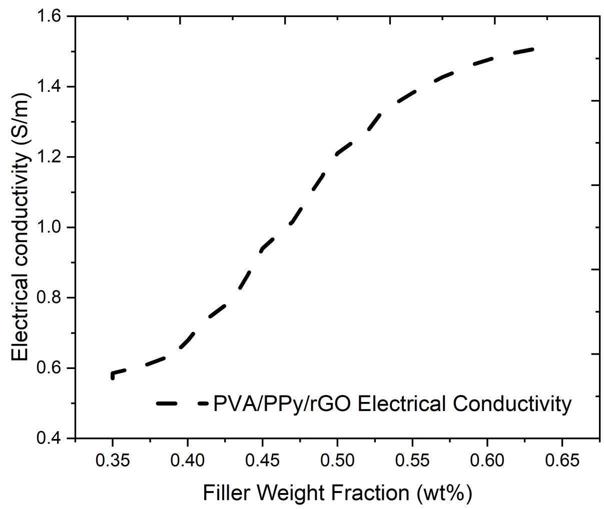

FIGURE 3. RGO/PVA/PPy nanocomposite electrical conductivity data.

saturation point begins at about 0.25 wt (%). At saturation, the electrical conductivity of the nanocomposite is no longer dependent on the filler content. Similar trend is also observed for PVA/PPy/rGO nanocomposite electrical conductivity data shown in Figure 3.

Possibly, the adjustment of the electrical conductivity of polymer nanocomposites is a route to the control and improvement in the storage capacity, cycling, charge, and discharge of electrodes [40]. Therefore, the study of the electrical conductivity of polymer nanocomposites is central to developing substantive electrodes for grid power applications and electric vehicle advantage.

In order to test the performances of the developed series-parallel polymer nanocomposite electrical conductivity model, the experimental data from the RGO/PVA/PPy and Gr-PPy nanocomposites, were engaged. Figure 4 shows the comparison of the developed series/parallel electrical conductivity model with the RGO/PVA/PPy nanocomposite

TABLE 1. Series-parallel model performances.

Nanocomposite	Parameters	Parameter values	Standard Error	Standard Error (per unit)	R^2	R^2-adj
RGO/PVA/PPy	$\rho(S/m)$ t $\mu(S/m)$ $\delta(p.u)$	0.41 3.81 20.12 3.51	0.034 0.43 14.284 0.426	0.08 0.114 0.71 0.12	0.995	0.994
Gr-PPy	$\rho(S/m)$ t $\mu(S/m)$ $\delta(p.u)$	21.00 3.00 9009.00 0.08	5.85 0.27 654.50 0.02	0.28 0.09 0.07 0.30	0.998	0.995

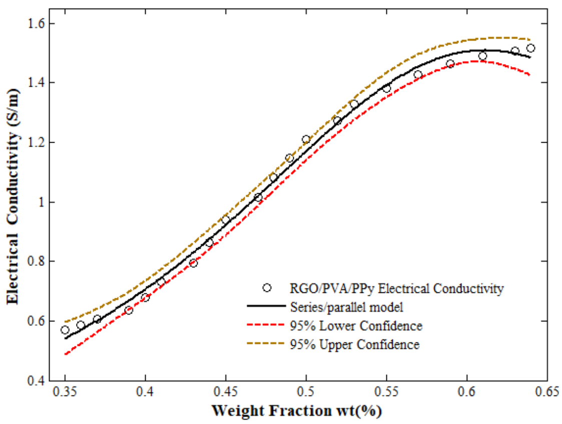

FIGURE 4. Series/parallel model for RGO/PVA/PPy electrical conductivity prediction.

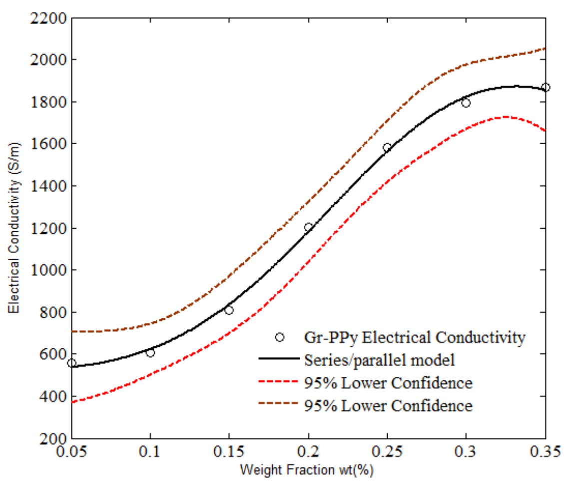

FIGURE 5. Series/parallel model for Gr-PPy nanocomposite electrical conductivity prediction.

electrical conductivity data. As shown in the figure, the developed series-parallel electrical conductivity model subjected to the electrical conductivity data of RGO/PVA/PPy nanocomposite displayed a good level of prediction of the conductivity of the nanocomposite. The agreement of the model with the experimental data confirms the model's capability to estimate the nanocomposite's conductivity.

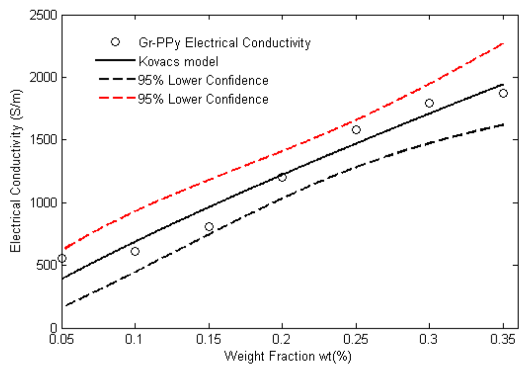

FIGURE 6. Comparison of Gr-PPy nanocomposite with Kovacs model.

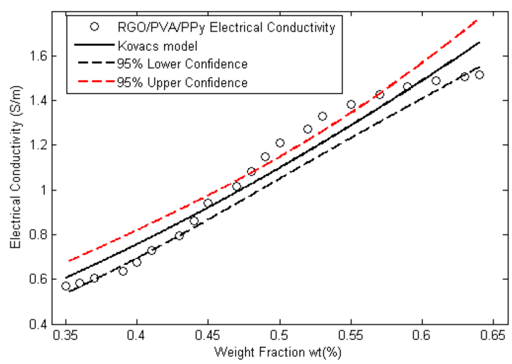

FIGURE 7. Comparison of RGO/PVA/PPy nanocomposite with Kovacs model.

Table 1 provides the model's calculated parameters for RGO/PVA/PPy nanocomposite. Also shown in Table 1, are the statistical accuracy of the model. Furthermore, Equation (20) provides the equivalent series/parallel electrical conductivity model for the RGO/PVA/PPy nanocomposite.

$$\sigma_{eff} = \frac{0.82\left((1-x+x^2)x^{-1}\right)^{3.81} + 20.12(1-x)^{3.81}}{1+3.51\left(\frac{1-x}{x}\right)^{3.81}} \tag{20}$$

Moreover, the applicability of the model is tested on the electrical conductivity of Gr-PPy nanocomposite, shown in Figure 2. From the results of the series-parallel electrical

TABLE 2. ANN model predictive results and performance for RGO/PVA/PPy nanocomposite.

Data points	1	3	5	7	9	11	13	15	17	19
Measured value (S/m)	0.57054	0.60583	0.67769	0.79387	0.93986	1.08249	1.21047	1.32998	1.42690	1.48900
Predicted error (%)	0.13300	0.54965	0.97242	0.01133	0.52561	0.27806	2.58329	2.40003	0.59570	0.12760
Mean (30 runs) (s/m)	0.58130	0.59440	0.69848	0.78479	0.92738	1.06353	1.18843	1.36015	1.44361	1.50295
Median (30 runs) (S/m)	0.57790	0.60580	0.68509	0.79389	0.93985	1.08457	1.21046	1.32997	1.42690	1.48900

TABLE 3. ANN model predictive results and performance Gr-PPy nanocomposite.

Data points	1	3	5	7	9	11	13
Measured value (S/m)	559.41	608.55	808.44	1201.88	1579.14	1791.14	1869.33
Predicted error (%)	0.31	1.39	0.0025	0.0091	0.029	0.23	1.73
Mean (30 runs) (S/m)	565.40	613.78	783.57	1238.57	1568.58	1799.95	1888.91
Median (30 runs) (S/m)	559.41	608.55	808.44	1201.88	1579.58	1791.14	1869.38

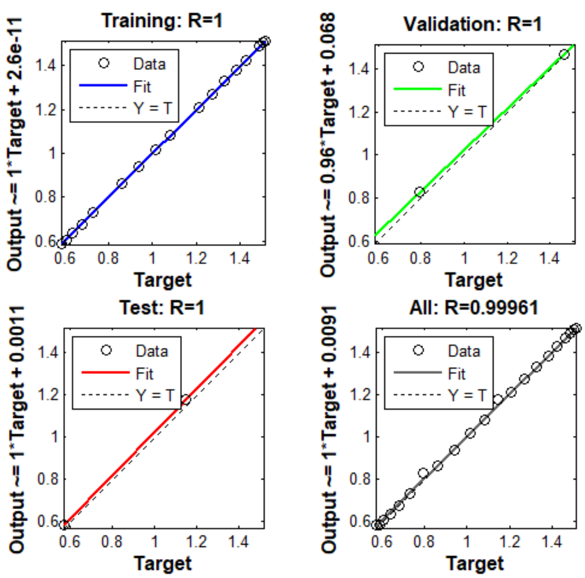

FIGURE 8. Model regression for RGO/PVA/PPy nanocomposite electrical conductivity data.

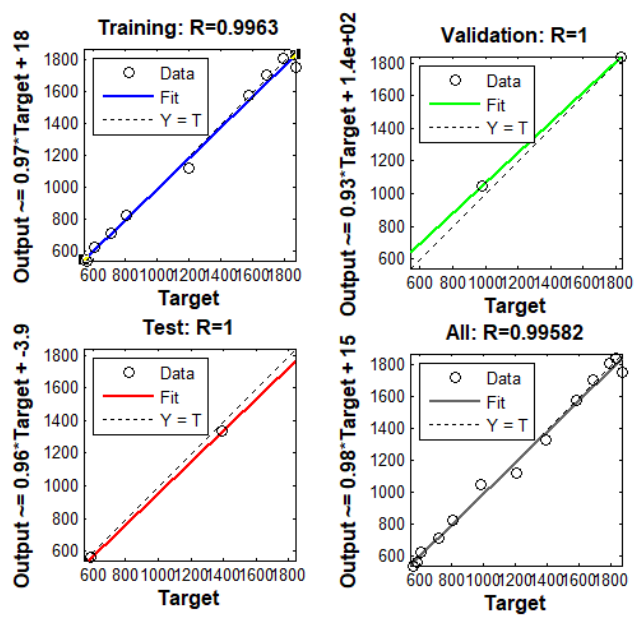

FIGURE 9. Model regression for Gr-PPy nanocomposite electrical conductivity data.

conductivity model, its capability is revealed as a suitable model that can be used to predict the electrical conductivity of multi-fillers and their nanocomposites. Without prior knowledge of electrical conductivity, the model is capable of predicting the conductivity of the additives and the fillers in nanocomposites. As shown in Figure 5, the developed model predicted the electrical conductivity of the Gr-PPy nanocomposite with high accuracy. Table 1 also provides the model's performance results. The per unit error showed that there is a minimal error in the calculated parameters of the model. Moreso, the statistical value of how close the conductivity data is to the regression line, evident the suitability of the model in quantifying and characterizing the electrical conductivity of polymer nanocomposites. From the results of the predicted electrical conductivity of the Gr-PPy nanocomposite, Equation (21), is as presented. An observation from the two electrical conductivity data is that the connecting exponent is within the same range of values. Hence, this model is suitable for calculating the transport properties of polymer nanocomposites.

$$\sigma_{eff} = \frac{41\left((1-x+x^2)x^{-1}\right)^{3.00} + 9009(1-x)^{3.00}}{1+0.08\left(\frac{1-x}{x}\right)^{3.00}} \tag{21}$$

TABLE 4. ANN model predictive results and performance for RGO/PVA/PPy nanocomposite.

Data points	1 (S/m)	2 (S/m)	3 (S/m)	4 (S/m)	5 (S/m)	6 (S/m)	7 (S/m)	8 (S/m)	9 (S/m)	10 (S/m)	11 (S/m)	12 (S/m)	13 (S/m)	14 (S/m)	15 (S/m)	16 (S/m)	17 (S/m)	18 (S/m)	19 (S/m)
	0.5779	0.5779	0.5859	0.6366	0.6933	0.7306	0.7970	0.8650	0.9348	1.0423	1.1398	1.1466	1.1174	1.2721	1.3299	1.3822	1.4267	1.4627	1.4889
	0.5855	0.5855	0.6183	0.6366	0.6781	0.7301	0.5961	0.6306	0.8508	1.0191	1.0866	1.1464	1.2039	1.2723	1.3118	1.3818	1.4272	1.4623	1.4892
	0.5865	0.5865	0.5339	0.6482	0.6882	0.7166	0.8235	0.8698	0.9739	1.0557	1.0846	1.1286	1.2496	1.3021	1.3064	1.3915	1.3041	1.4179	1.5244
	0.5853	0.5853	0.6058	0.5091	0.6382	0.7305	0.7939	0.8354	0.9399	1.0156	1.0825	1.1467	1.2105	1.2725	1.3300	1.3821	1.4427	1.4625	1.4621
	0.5779	0.5779	0.9006	0.9546	0.8146	0.7305	0.8288	0.8632	0.9399	1.0156	1.0825	1.1467	1.2105	1.2725	1.3300	1.3956	1.4269	1.4625	1.4890
	0.8516	0.8516	1.0536	0.6367	0.6899	0.7306	0.7905	0.8583	0.9396	1.0145	1.0861	1.1482	1.2173	1.2712	1.3287	1.3821	1.4268	1.4626	1.4890
	0.3058	0.3058	0.0643	0.4724	0.6935	0.8444	0.8896	0.8826	0.8426	0.8307	0.8444	0.8767	0.9658	1.3833	1.6332	1.0428	1.3532	1.6847	1.7728
	0.8263	0.8262	0.6058	0.6366	0.6777	0.7013	0.7939	0.8632	0.9399	1.0156	1.0825	1.1467	1.2105	0.9865	0.8615	1.3821	1.4269	1.4625	1.4890
	0.5778	0.5778	0.5849	0.6390	0.6844	0.7868	0.8322	0.8695	0.9521	1.0120	1.0877	1.1439	1.2053	1.2864	1.3299	1.3822	1.4268	1.4628	1.4888
	0.5820	0.5820	0.3035	0.6471	0.6907	0.7220	0.7965	0.8335	0.9684	1.0739	1.0790	1.1151	1.2172	1.2821	1.3038	1.3774	1.4633	1.4925	1.4791
	0.5784	0.5784	0.6062	0.5065	0.6766	0.7305	0.7939	0.8299	0.9391	1.0292	1.0846	1.1504	1.2164	1.2740	1.3291	1.3821	1.31935	1.3608	1.4890
	0.5850	0.5850	0.8264	0.6605	0.6938	0.7213	0.8044	0.8210	0.8528	0.9935	1.0974	1.1325	1.18860	1.2900	1.3612	1.3561	1.4053	1.4709	1.5001
	0.5853	0.5853	0.6058	0.6366	0.6777	0.7122	0.7939	0.8332	0.9399	1.0491	1.1075	1.1467	1.2105	1.2725	1.3300	1.3821	1.4269	1.4625	1.3107
	0.5779	0.5779	0.4576	0.5829	0.6777	0.7305	0.7939	0.8219	0.8896	1.0156	1.1021	1.1467	1.2105	1.2725	1.3300	1.3821	1.4269	1.4589	1.4890
	0.5779	0.5779	0.6058	0.6366	0.6977	0.7305	0.7939	0.8632	0.9353	1.0156	1.1358	1.1467	1.05303	1.2057	1.3300	1.3821	1.4269	1.4625	1.4890
	0.5717	0.5717	0.6187	0.6360	0.6844	0.7271	0.8049	0.8502	0.9274	1.0163	1.0841	1.1376	1.2902	1.6329	1.8041	1.3821	1.4097	1.4775	1.4873
	0.5714	0.5714	1.0250	0.6072	0.6897	0.7392	0.6885	0.7481	1.0063	1.1184	1.1007	1.1077	1.1443	1.2702	1.3484	1.3839	1.3136	1.3675	1.4784
	0.5705	0.5705	0.6058	0.6467	0.6777	0.7305	0.7939	0.8632	0.9399	1.0156	1.0825	1.1467	1.2966	1.6215	1.7959	1.3821	1.4009	1.4625	1.4890
	0.5853	0.5853	0.6058	0.6366	0.6777	0.7160	0.8352	0.8632	0.9399	1.0530	1.0825	1.1279	1.2105	1.2725	1.3210	1.3821	1.4269	1.4625	1.4499
	0.5705	0.5705	0.6058	0.6366	0.6664	0.6895	0.7939	0.8631	0.9398	0.9860	1.0825	1.1467	1.2105	1.2871	1.3299	1.3820	1.4269	1.4625	1.4890
	0.5705	0.5705	0.2025	0.3800	0.6298	0.7305	0.7939	0.8632	0.9399	1.0325	1.1065	1.1467	1.2105	1.2725	1.3299	1.3821	1.4269	1.4625	1.4889
	0.5725	0.5725	0.5886	0.6696	0.6931	0.7113	0.8286	0.8649	0.9206	1.0144	1.0897	1.1393	1.2390	1.2941	1.3250	1.7505	1.4263	1.3340	1.4856
	0.6614	0.6614	0.5892	0.6201	0.6929	0.7477	0.7965	0.8249	0.9125	1.02049	1.0846	1.1237	1.1921	1.2773	1.3252	1.4155	1.4394	1.4661	1.4930
	0.5779	0.5779	0.4979	0.5945	0.6777	0.7305	0.7939	0.8326	0.9399	1.0156	1.0542	1.1161	1.2105	1.2725	1.3300	1.3821	1.4269	1.4625	1.4890
	0.5779	0.5779	0.9033	0.6366	0.7378	0.8025	0.8352	0.8632	0.9399	1.0156	1.0825	1.1118	1.1534	1.2725	1.3300	1.3821	1.4269	1.4625	1.4890
	0.5700	0.5700	0.6100	0.6105	0.6790	0.7292	0.7954	0.8356	0.9200	1.0174	1.0867	1.1371	1.2019	1.2769	1.3243	1.3832	1.4236	1.4654	1.4861
	0.5779	0.5779	0.6058	0.6366	0.6777	0.7305	0.7939	0.84546	1.0058	1.0156	0.9146	0.9172	0.9899	1.2725	1.3300	1.3821	1.4269	1.4625	1.6149
	0.5779	0.5779	0.6605	0.6366	0.6979	0.73054	0.7939	0.8632	1.0454	1.01565	0.9619	1.0748	1.2105	1.2725	1.3300	1.3937	1.4269	1.4625	1.4890
	0.5818	0.5818	0.6182	0.6316	0.6858	0.7277	0.7983	0.8372	0.9461	1.0457	1.0946	1.1166	1.1634	1.2704	1.3315	1.3851	1.3902	1.4614	1.4933
	0.3426	0.3426	0.1388	0.7598	1.0149	1.1550	0.4761	0.4351	0.6595	0.7958	0.8153	0.9288	1.2429	1.4305	1.4949	2.0098	2.3863	2.22039	1.7161
Mean	0.5814	0.5814	0.5945	0.6260	0.6985	0.7482	0.7848	0.8264	0.9274	1.0125	1.0635	1.1116	1.1884	1.2971	1.3602	1.4054	1.4436	1.4847	1.5030
Median	0.5779	0.5779	0.6058	0.6366	0.6851	0.7305	0.7939	0.8543	0.9399	1.0156	1.0846	1.1384	1.2105	1.2725	1.3300	1.3821	1.4269	1.4625	1.4890

TABLE 5. ANN model predictive results and performance for RGO/PVA/PPy nanocomposite.

Data points	1 (S/m)	2 (S/m)	3 (S/m)	4 (S/m)	5 (S/m)	6 (S/m)	7 (S/m)	8 (S/m)	9 (S/m)	10 (S/m)	11 (S/m)	12 (S/m)	13 (S/m)
	559.41	628.89	608.55	618.27	808.44	979.51	1201.88	1388.34	1608.45	1687.15	1791.14	1906.84	1869.33
	559.45	584.37	608.53	830.20	898.43	979.51	1201.88	1388.34	1579.58	1687.14	1626.29	2135.41	1869.34
	550.98	549.42	456.71	79.28	487.79	1116.39	1130.28	1371.87	1590.82	1696.37	1815.55	2156.12	1872.47
	559.43	584.31	608.54	915.49	808.44	653.91	1201.88	1388.34	1563.59	1687.15	1791.14	1832.94	1969.64
	559.37	851.93	617.35	605.58	836.15	993.00	1200.86	1384.63	1579.25	1686.76	2015.79	1832.88	1387.30
	604.30	584.37	736.66	42.48	327.59	998.30	1208.58	1389.62	1582.79	1681.31	1832.64	1850.71	1800.39
	559.41	532.94	608.55	714.66	808.44	979.51	1119.84	1479.58	1579.58	1687.15	1791.14	1832.94	1537.98
	483.69	583.29	127.67	714.64	812.80	659.37	1203.31	1383.77	1664.82	1872.98	1780.78	1833.72	1878.34
	537.34	601.34	620.69	658.30	793.76	975.75	1201.73	913.99	1253.83	1684.99	1618.12	2150.05	2042.38
	−30.02	−228.83	608.60	714.88	808.77	986.43	1200.64	1387.09	1599.55	1687.85	1792.72	1832.77	1870.34
	324.44	584.30	608.55	714.66	808.44	979.51	1201.88	1388.34	1593.97	1826.08	1791.14	1832.94	1955.54
	766.91	653.10	680.95	671.03	729.79	979.61	985.03	1636.97	1771.33	1688.93	1721.80	1854.15	1873.78
	559.39	451.68	608.56	714.65	808.45	979.54	1201.88	1388.34	1547.47	1779.11	2336.55	1832.96	1869.44
	563.02	572.11	614.22	713.91	806.74	979.43	1382.39	1388.29	1581.87	1913.89	2037.60	1832.94	1929.16
	868.11	584.30	608.55	714.66	839.79	979.51	1201.88	1388.34	1579.58	1687.15	1383.82	1832.94	2372.88
	559.41	584.30	608.55	714.66	922.97	1084.72	1201.88	1388.34	1562.00	1687.15	1791.14	1832.94	2080.86
	796.89	584.30	608.55	714.66	808.44	979.51	1556.92	1509.40	1579.58	1687.15	1791.14	2054.98	1869.33
	689.88	588.39	980.43	752.654	793.56	935.89	1204.84	1377.94	1540.58	1691.71	1786.58	2028.51	1827.53
	559.41	560.74	608.55	714.66	889.29	979.51	438.25	1388.34	1579.58	1567.25	1791.14	1832.94	1869.33
	559.41	531.20	608.55	714.66	808.44	979.51	1581.26	1655.35	1595.50	1687.15	1791.14	1832.94	1869.33
	829.58	584.79	910.69	725.55	703.42	970.58	1288.55	1396.38	1494.68	1621.25	1785.88	1830.38	1782.84
	572.21	541.13	657.15	722.36	704.60	1008.07	1326.45	1366.54	1572.89	1574.29	1525.65	1661.34	1854.19
	559.42	584.32	523.65	714.66	808.44	922.45	1201.88	1430.70	1579.58	1687.15	1791.14	1832.94	2039.06
	559.48	584.27	290.92	714.66	786.02	979.47	1627.32	1389.91	1577.11	1686.74	1791.84	2351.78	1869.43
	494.81	737.20	608.55	714.66	808.44	979.51	1201.88	1284.69	1579.58	1687.15	1791.14	2176.24	1869.33
	559.41	584.30	608.55	714.66	816.33	979.51	1413.42	1388.34	1579.58	2071.46	1791.14	1832.94	2143.64
	559.41	584.30	608.56	602.03	808.44	979.51	1201.88	1488.22	1579.58	1687.15	1735.64	1832.94	2130.34
	526.79	662.80	678.98	709.63	837.90	980.37	1453.34	1541.60	1550.11	1717.35	2108.28	1835.15	1785.80
	510.94	460.22	730.32	728.15	750.62	982.01	1438.94	1136.85	1409.24	1691.28	1795.77	1885.55	1775.16
	599.62	591.41	658.83	701.47	876.35	976.33	1176.43	1414.14	1601.40	1706.88	1804.73	2388.12	1802.96
Mean	565.40	562.71	613.78	669.39	783.57	964.54	1238.57	1394.09	1568.58	1713.50	1799.95	1925.33	1888.91
Median	559.41	584.30	608.55	714.66	808.44	979.51	1201.88	1388.34	1579.58	1687.15	1791.14	1832.95	1869.38

To further exemplify the suitability of the proposed model, the previous model shown in Equation 4 was applied to predict the electrical conductivity of Gr-PPy and RGO/PVA/PPy nanocomposites. As shown in Figures 6&7, the model inaccurately predicted the electrical conductivity data of the nanocomposites. Even though the coefficient of determination of the Kovacs model for the two nanocomposites is reasonable (0.9547 for Gr-PPy and 0.9477 for RGO/PVA/PPy), nevertheless, the model regression is inconsistent with the study data [41]. It is expected that models for forecasting the characteristics of hybrid materials, must properly define the nature of each composited material and their interactive behavior. These conditions, lacking in the Kovacs model, might be the underlying factor responsible for the model's inaccuracy. However, the model's modification might result in a useful model for predicting the electrical conductivity of polymer nanocomposite.

A. ANN RESULTS

A vital factor to consider before performing artificial neural network modeling is the volume of the data points the model is to train. Due to the cost and time required to perform the experimentation of the electrical conductivity of polymer nanocomposites, few samples in the range of ten are usually produced. Therefore, optimizing the experimentation results would be helpful for better performance and accuracy of the model. Afterward, the experimental data must be subjected to three process divisions: training, validation, and testing [42]. In order to monitor the performances of the model, the regression plots of the model were observed for different weights and biases of the neural network. The number of neurons engaged for the fitting, are: 13 (hidden), and 1 (output). A 70/100, 15/100, and 15/100 training, validation, and testing ratios, were chosen for the data. The training time is approximately 1.0 seconds. As shown in Figures 8&9, the suitability of the model for the trained electrical conductivity data is presented. From the regression plots, the model's performance is observed, and its appropriateness for the data prediction is indisputable. For both nanocomposite electrical conductivity, the model showed a R^2 coefficient performances in the range of 0.999 and 1.000 for validation and training.

Furthermore, the comparison of the measured electrical conductivity of the nanocomposites with the ANN predicted values, are as shown in Tables 2&3. By considering the small and insignificant percentage error recorded for the predicted values, the ANN model can be said to have shown a high percentage of accuracy and agreement between the measured and the predicted values. Hence, the ANN model can be classified as an efficient modeling method for the prediction of the electrical conductivity of polymer nanocomposites [18], [43], [44]. Therefore, in the absence of experimentation, the ANN model is a vital tool for the prediction of the properties of polymer nanocomposites. In summary, an artificial neural network has a short-time training process and can detect complex nonlinear relationships between dependent and independent variables. In addition, artificial neural networks can effectively predict an output from arrays of input data without prior knowledge of the data's hidden relationships. Compared with conceptualized models, artificial neural network model involves a simple modeling process and is usually viewed as a nonlinear generalization of logistic regression [45]. Referring to the discussed performance indicators for both models, it can be inferred that the developed classical equation and the ANN model, are beneficial for the characterization of electrical conductivity of polymer nanocomposites. Appendix presents the full-length prediction of the artificial neural network for thirty runs.

V. CONCLUSION

This study developed a unique and versatile series-parallel model for the prediction of the electrical conductivity of polymer nanocomposites. The consideration of the series-parallel model developed is based on the connectivity of the filler and the polymer. The connectivity can be defined in terms of tunneling and contact resistance between the hybrid materials. The coefficients of determination of the series-parallel mode, are: 0.995 for RGO/PVA/PPy and 0.998 for Gr-PPy nanocomposites. From the results of the model, it can be concluded that it is suitable for calculating and predicting the transport properties of polymer nanocomposites. Furthermore, the study considered the applicability of artificial neural networks in the nanocomposite's electrical conductivity prediction. The average calculated predicted errors of the ANN for RGO/PVA/PPy and Gr-PPy nanocomposites, are 0.82% and 0.53%. The proposed artificial neural network model is accurate and in agreement with the experimental data. By considering the accuracy of the models for the computational study of the electrical conductivity of polymer nanocomposites, they are therefore recommended for the investigation of the transport properties of all types of polymer nanocomposites.

APPENDIX

See Tables 4 and 5.

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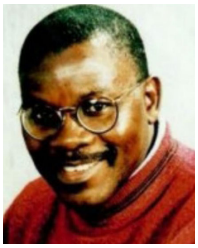

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