Enhancement of electronic protection to reduce e-waste

Yadollah Abdollahi a, b, *, Suhana Binti Mohd Said a, Nor Asrina Sairi c, Mohd Faizul Bin Mohd Sabri d, Azmi Zakaria a, Ebrahim Abouzari-lotf e, Masoumeh Dorraj b, Raba‘ah Syahidah Aziz f

a Department of Electrical Engineering, Faculty of Engineering, University of Malaya, 50603 Kuala Lumpur, Malaysia
b Material Synthesis and Characterization Laboratory, Institute of Advanced Technology, Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia
c Department of Chemistry, Faculty of Science, University of Malaya, 50603 Kuala Lumpur, Malaysia
d Department of Mechanical Engineering, Faculty of Engineering, 50603 Kuala Lumpur, Malaysia
e Institute of Hydrogen Economy, Energy Research Alliance, International Campus, Universiti Teknologi Malaysia, 54100 Kuala Lumpur, Malaysia
f Department of Physics, Faculty of Science, Universiti Putra Malaysia, UPM, 43400 Serdang, Selangor, Malaysia

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ABSTRACT

The electronic devices turn to e-waste due to their insufficient electrical protection which is provided by a ceramic core varistor. The ceramic consists of the surrounded ZnO grains of melted an additives layer. The layer is origin of the quality of the protection. To enhance the quality and consequently prevent e-waste, the fabrication of the varistor was modeled by artificial neural network. The model predicted the optimized condition that was experimentally fabricated and electrically characterized. The results confirmed the model predictability. In conclusion, the optimization which has industrial scales up potential warranties the electronic protection that controls the global e-waste.

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Introduction

Due to the short useful lifetime of electronic products they turn to e-waste which originates pollution problem for hydrosphere, biosphere even atmosphere [1–3]. Generally the harmful materials of estimated global e-waste which is about 35 to 50 million tons per year includes nano-materials that contains lead, chromium, mercury, cadmium and arsenic into environment [4–12]. To control the e-waste, the existed current methods are including reusing, refurbished and recycling process of second-hand electronics. Recycling process covers only 23% of the e-waste which may not be fully recovered even the amount of the hazardous materials [13]. In addition, the reusing and refurbished processes are unable to even postpone the old technology any more while the generation rate of very short lifetime electronic devices is quite high around the world [14].

On the other hand, the lifetime could be improved by high quality electrical protection from common generated overvoltage including lightning strikes, power outages tripped circuits, power transitions, power malfunctions, electromagnetic pulses and inductive spikes in the associated circuit [15]. The overvoltage damages the electrical devices which designed to operate at normal voltages. Currently, the protection as a preventive action is carried out by a voltage-limiting device such as varistor which is associated in parallel of electronics into the electrical circuit. It means the varistor has presented high resistance ohmic behavior within the operating normal voltage that the normal electrical current never flows through the associated varistors [16]. On the other hand, the varistor changes into non-ohmic behavior at the overvoltage and allows the current to flow through itself. In this way, the varistor diverts the overvoltage safely from the device at certain threshold voltage [17,18]. However, most discarded electronic appliances were damaged due to the overvoltage that must be deflected by the associated varistors. It is obvious that the varistors unable to protect the devices during surge due to their insufficient non-ohmic behavior which is originated from the used ceramic core.

The used ceramic in the varistors has been made of n-type semiconductor such as zinc oxide (ZnO) and other metal oxides as
additive [19,20]. Since, the microstructure of the ceramic is made of ZnO grains that is surrounded by narrow boundaries of melted additives as segregation layer [21,22]. The additives are included Bi$_2$O$_3$, TiO$_2$, Co$_3$O$_4$, Mn$_3$O$_4$, Sb$_2$O$_3$, V$_2$O$_5$, and Al$_2$O$_3$ which initially mixed with a large amount of ZnO by physical or chemical methods [23–25]. Thereafter the mixed final powder is compacted and heated to occupy the grains boundaries [26–31]. In fact, the melting points of the additives are less than the melting point of ZnO by reason of they are melted to fill up the boundaries [32,33]. In the layer, Bi$_2$O$_3$ is used as former and other additives are subordinate which often included improving the ceramic electrical properties [17,33–35]. For instance, TiO$_2$ prevents the vaporization of Bi$_2$O$_3$ and also facilitates ZnO grain growth; Sb$_2$O$_3$ stabilizes the electrical properties and diminishes the leakage current of the varistor during performance [17,29,30,36]. The other additive such as Co$_3$O$_4$, Mn$_3$O$_4$, and Al$_2$O$_3$ are involved in the formation of interfacial states which contribute to the highly non-ohmic property (non-linear property) [18,37]. Obviously, some of the additives make the property while others improve it. As a result, to enhance the non-ohmic property, the additives should be optimized in the starting powder of the ceramic.

In the case of the optimization, the method of ‘one variable at a time’ has been widely used by varying one of the additives while other parameters are kept constant while the additives are not completely independent; it affects the electrical property of the ceramic [36–39]. Moreover, the number of experiments is quite high due to the variety of the additives which entail time consumption and possible misinterpretation of the related results. More than that, the different reactions including formation and decomposition of spinels phase, kinetic of ZnO grain growth, densification and evaporation of additives during the varistor fabrication add to the complexity. Likewise, the importance of the variable which uses to determine the level of initial additives is impossible for the methods.

On the other side, the multivariate methods such as response surface methodology (RSM) and artificial neural networks (ANNs) contemplate the simultaneous effect of the input variables on the output free of the mentioned complexity [40–44]. However, RSM involves the complicated statistical calculation of fitting process as well as the regression analysis while ANNs are free of the mathematic functionalization [16,42,45,46]. ANNs have been successfully used for modeling of productive processes such as photodegradation of many environmental organic pollutants including ethylene-diamine-tetra-acetic acid [47], nitrogen oxides [48], nitritolactiacid [49], C1. Basic Red 46 [50], 2,4-dihydroxybenzoic acid [51] and 4-nitrophenol [52]. To the best of our knowledge, no study has shown the modeling of the additives as input variables of the ceramic fabrication.

In this work, ANN was used to model the fabrication of 26 ceramic cores which used to prepare the same numbers of ZnO based low voltage varistor. The amounts of the mentioned additives were selected as input effective variables while the calculated non-linear coefficients (alpha = α, from $I = KV$) of the experimental varistor's electrical characterization were used as outputs (responses). The modeling was carried out by four particular training algorithm programs which included Quick propagation (QP), Incremental back-propagation (IBP), Batch back-propagation (BBP) and Levenberg–Marquardt (LM) back-propagation algorithm [53,54]. Thereafter, the produced models of the algorithms are compared to find the optimized final model by the root means squared error (RMSE), the coefficient of determination ($R^2$) and the percentage of absolute average deviation (AAD) of the obtained models for each algorithm. The final model was used for navigation of the fabrication to determine the narrow levels and importance of the additives as well as predicting the points of the additives that maximize the non-linearity of the varistors. The predicted condition was experimentally prepared and electrically validated to determine the protectiveness and sustainability of the optimized varistor. The result of the validation was quite close to the predicted condition.

**Experiment**

**Materials and methods**

The used chemicals were included ZnO (99.99%), Bi$_2$O$_3$ (99.975%), TiO$_2$ (99.9%), Co$_3$O$_4$ (99.7%), Sb$_2$O$_3$ (99.6%), Mn$_3$O$_4$ (98%) and Al(NO$_3$)$_3$ (100%) which provided from Alfa Aesar for preparation the ceramic starting powder. For fabricate a varistor, the appropriate amount of each above chemicals was mixed and grounded by dry form and then wet ball milled for 24 h to prepare initial homogenous mixed powder. The mixed powder was overnight dried by a hot oven at 100 °C then it was pressed into pellet forms (10 mm in diameter and 0.70 mm thickness) at 200 Mpa by a uniaxial presser machine. The pellet was sintered for holding time of 1 h at 1260 °C while the heating and cooling rate were 5 °C/min by a box furnace (CMTS model HTS 1400) [40]. Thereafter, the both sides of the sintered pellet as ceramic core of the varistor were painted by silver electrodes to scan DC current–voltage. The scan was carried out from 0 to 100 volts in step size of 2.5 V by aKeithly source-meter 2400. The current–voltage (I–V) was used to calculate the current density, $J$ (mA/cm$^2$) and electrical field, $E$ (V/mm) where ‘mm’ is the thickness of pellets and ‘cm$^2$’ is the surface of the painted silver electrodes. The “$E$” was plotted vs.“$J$” to calculate the alpha of the varistor at different values according to the following equation [55]:

$$\text{Alpha} = \frac{[\log(J_2 - \log(J_1))]}{[\log(E_2 - \log(E_1)]} \quad (1)$$

where $J_1 = 0.1$, $J_2 = 1$ mA/cm$^2$, $E_1$ and $E_2$ were measured at $J_1$ and $J_2$, respectively. The process was carried out for 26 varistors with different mol% of starting powders in their ceramic core (Table 1). The data of the varistors were randomly split up into two sets as training and testing data sets (Table 1) using the option available in NeuralPower software version 2.5 [56,57]. The training and testing data were used to compute and ensure robustness of the network parameters, respectively.

**Theory of the modeling**

ANNs are semi-empirical modeling methods which use the actual processing condition and corresponding responses to govern a network to avoid of the process complexity. The network consists of input, hidden and output layers which are made of appropriate connected units (nodes). The nodes are simple artificial neurons which mimic a biological neural network. The nodes of input layer are the effective variables and in output layer is the responses. In the hidden layer, the number of nodes is determined by learning process [58,59]. In the network, the nodes are connected by multilayer normal feed-forward or feed-back connection formula [53]. To qualify the network, the input layer acts as distributor and sends data via the weights to the nodes of second layer (hidden layer) [60]. The weighted data is saved as processing nodes in the hidden layer and then transferred to the output layer by particular transferred function [61,62]. Therefore, the qualified data are passed into the input layer, propagated to hidden layer and then transfers into the output layer of the network by iterative procedure [63]. The iteration is an act of repeating a process to approach a desire result. After appeared the first input–output iteration result, the second period is processed and so on. The network changes the weights in order to reduce the
difference between actual and network's predicted responses at each iteration. The results of iteration are used as starting point of next iteration. For example, when the results of last iteration become almost equal to the results of previous iteration, the process will be terminated. The iteration process is continued by self-similarity method (Eq. (2)) [63].

$$S(B) = \sum_{i=1}^{m} [y_i - f(x_i, \beta)]^2$$

where \( m \) is an empirical data pairs of independent and dependent variables such as \( (x_i, y_i) \) and \( f(x_i, \beta) \) is the model curve. In self-similarity process, the \( \beta \) parameter of \( f(x_i, \beta) \) is optimized by minimizing the root mean squared error (RMSE). As a result, the main aim of the learning process is to find the weights for minimizing the RMSE which is obtained from difference between network prediction and actual responses (Eq. (3)).

$$\text{RMSE} = \left( \frac{1}{n} \sum_{i=1}^{n} (y_i - y_{di})^2 \right)^{1/2}$$

where \( n \) is number of the points, \( y_i \) is the predicted values and \( y_{di} \) is the actual values.

In this case, the number of nodes in the hidden layer was obtained by trial and error learning calculation which was examined from one to 15 nodes. The learning process was initially started with one node in the hidden layer to obtain network (architecture) with 6 input nodes, 1 node in hidden and 1 node in output layer by QP algorithm. The nodes in the input and output layers are kept constant during the process while number of the nodes in the hidden layers were varied up to 15. The examination of each node is repeated for ten times to avoid the random correlation due to the random initialization of the weights. Among the repeated examination node number, the architecture with the lowest RMSE is selected to compare with other architectures. Therefore, 15 architectures are obtained at the end of the learning process for QP algorithm. As a result of the learning process, the architecture with minimum RMSE is selected as a final topology for the QP. The similar process is carried out for IBP, BBP and LM algorithms to obtain 4 topology with minimum RMSE [53]. To obtain a model for the fabrication, the 4 final topologies are also compared by their \( R^2 \)-squared (\( R^2 \)) and AAD were calculated as the following equations:

$$R^2 = 1 - \frac{\sum_{i=1}^{n} (y_i - \hat{y}_i)^2}{\sum_{i=1}^{n} (y_{di} - \hat{y}_{di})^2}$$

$$\text{AAD} = \frac{\sum_{i=1}^{n} (|y_i - \hat{y}_i|/\hat{y}_i)}{n}$$

where \( n \) is the number of points, \( y_i \) is the predicted value, \( \hat{y}_i \) is the actual value, and \( y_{di} \) is the average of the actual values. Therefore, the appropriate topologies were determined by minimum RMSE and AAD while their \( R^2 \) is at maximum value. The model is used to obtain the importance and optimum narrow level of the additives in the initial powder. In addition, the model is able to predict the optimum values of the additives to achieve the maximum alpha value.

**Results and discussion**

**Model selection**

Fig. 1 illustrates the RMSE versus node number of the hidden layer which were obtained by learning process. Each node number presents the hidden layer of the architecture of QP, IBP, IBP, and LM algorithms. As shown, the minimum RMSE belonged to architecture with hidden layer of 10, 6, 14 and 4 nodes for QP, BBP, IBP and LM algorithms, respectively. Therefore, the candidate topologies for the model were QP-6-10-1, BBP-6-6-1, IBP-6-14-1 and LM-6-4-1 (algorithm-input layer, hidden layer-output layer). Among the topologies, QP-6-10-1 has presented the lowest RMSE.
however the topologies were evaluated for more accuracy by AAD and R².

Fig. 2 demonstrated the calculated AAD (Eq. (6)) results of selected topologies such as QP-6-10-1, BBP-6-6-1, IBP-6-14-1 and LM-6-4-1. As observed, topology of QP-6-10-1 exhibited the minimum AAD. In this case, the QP-6-10-1 was more effective than BBP-6-6-1, IBP-6-14-1 and LM-6-4-1 topologies.

In addition, Fig. 3 shows the scatter plots predicted alpha versus actual alpha of training data set to exhibit the R² of the QP-6-10-1, BBP-6-6-1, IBP-6-14-1 and LM-6-4-1. As shown, the predicted values of QP-6-10-1 topology was so well fitted to the actual values (R² = 0.9997). Moreover, the R² of these topologies in the testing date sets confirmed these results (Fig. 4). As shown, the R² (0.973) was quit high that depicted the qualification of QP-6-10-1 topology. Therefore, the QP-6-10-1 topology was considered as efficient final model for the ceramic fabrication (Fig. 5). The model has been applied for navigation of the
fabrication to obtain the importance and optimized level of the additives in the starting power. Finally, the model predicted the ceramic’s starting powder to achieve the maximum non-linearity \((\alpha)\) for the varistor.

**Model applications**

**Importance determination**

Importance shows the relative effect (%) of the input variables on the output. Fig. 6 shows the relative importance of \(\text{Bi}_2\text{O}_3\), \(\text{TiO}_2\), \(\text{Co}_3\text{O}_4\), \(\text{Mn}_2\text{O}_3\), \(\text{Sb}_2\text{O}_3\) and \(\text{Al}_2\text{O}_3\) in initial powder of the ceramic at optimum condition for selected model (QP-6-10-1). As observed, the relative importance was \(\text{TiO}_2 > \text{Bi}_2\text{O}_3 > \text{Mn}_2\text{O}_3 > \text{Co}_3\text{O}_4 > \text{Sb}_2\text{O}_3 > \text{Al}_2\text{O}_3\). As a result, the selected additives variables were confirmed as effective input for the ceramic fabrication and none of

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Fig. 4. The scatter plots of the predicted vs. actual alpha for testing data set to show the \(R^2\) of QP-6-10-1, BBP-6-6-1, IBP-6-14-1 and LM-6-4-1 as selected topologies.

Fig. 5. The QP-6-10-1 model structure of ZnO based low voltage varistor fabrication, the model consists of 6 variables in input layer, 10 nodes in hidden layer and 1 response (alpha) in output layer, the variables are \(\text{Bi}_2\text{O}_3\), \(\text{TiO}_2\), \(\text{Co}_3\text{O}_4\), \(\text{Mn}_2\text{O}_3\), \(\text{Sb}_2\text{O}_3\), \(\text{V}_2\text{O}_5\), and \(\text{Al}_2\text{O}_3\) and bias shifts the space of the non-linearity properties [64].

Fig. 6. The relative importance of \(\text{TiO}_2\) (33%), \(\text{Bi}_2\text{O}_3\) (22%), \(\text{Mn}_2\text{O}_3\) (17%), \(\text{Co}_3\text{O}_4\) (11%), \(\text{Sb}_2\text{O}_3\) (9%) and \(\text{Al}_2\text{O}_3\) (7%) as the used additives in initial powder of ceramic core for ZnO based low voltage varistor.
them was neglect able in this work. Among them, TiO$_2$ as an enhancement has presented the highest importance.

**Level optimization**

The wide levels of the additives in the starting power were selected according to previous works which were carried out by traditional methods. Therefore, the levels were re-designed and optimized by the validated model (QP-5-10-1). For this purpose, the model simulated the effect of two additives on the alpha simultaneously without further requirement of mathematic function and equation knowledge. The important simulated effects have been presented as 3D plots by Fig. 7 while the other plots are indicated by supplementary materials. Fig. 7 demonstrate the surface of the additives’ effect on the alpha of the varistor therefore, the optimized value of the additives were presented by the particular levels in mol%. The levels were Bi$_2$O$_3$ (0.85–1.35), TiO$_2$ (0.8–1.55), Mn$_2$O$_3$ (0.4–0.45), Co$_3$O$_4$ (0.4–0.8), Sb$_2$O$_3$ (0.4–0.5) and Al$_2$O$_3$ (0.00088–0.00138) at optimum condition. The levels were used to predict the optimum value additives’ points that maximized the alpha.

**The quality of the optimized varistor**

**Protectiveness**

The protectiveness of the varistor is expressed by the value of non-linear property (Eq. (1)). For this purpose, the model was used

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**Fig. 7.** The 3D plots of simultaneous effect of two additives on the alpha, the red surface response is the desirable alpha and blue color shows the lowest values of the alpha. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
to predict the optimum condition in the optimized levels (above section) for fabrication of 5 varistors (Table 2). As the table shows, optimum point value of the additives and the related alpha for each suggested varistors. The fabricated processes including preparation of the starting powder, pressing, sintering and electroding were carried out for final varistors in the laboratory. The electrical characterization of the varistors was carried out to calculate the alpha (Eq. [1]) which indicated in Table 2. As shown, the actual alphas with acceptable error were very close to the model predicted alpha. The high values of the alphas have presented the high quality protective of the varistors.

Stability

Stability has been exhibited by measured recovery alpha after removing the overvoltage. In this case, changing of $J$–$E$ graph after passing several times overvoltage. Fig. 8 shows the $E$–$J$ plot of first to fourth try scanned electrical characteristic from 0 to 100 of the final predicted varistor (varistor 3). As the point values of the different scanning shown, there is no changing in the non-linear property which expressed the great stability of varistor after fourth shocking. Moreover, the leakage current was 0.013 mA/cm² which confirmed the stability of the varistor. As observed the sustainability was quit significant after fourth over voltage.

Conclusion

Electronic ceramic is used as core of varistor devices which have been widely applied to protect the electronic and electronic devices. The ceramic is made of n-type ZnO which coated by a few number of additives such as Bi₂O₃, TiO₂, CoOₓ, Mn₂O₃, Sb₂O₃ and Al₂O₃. The additives are melted during sintering process to occupy the ZnO grains boundaries in ceramic microstructure. The melted additives have presented the non-linear property that is directly proportional to the varistor quality for unwanted voltage protection. The amount of the additives affects the non-linearity and consequently the quality of the varistor devices. In this work the amounts of the additives were modeled as effective variables for maximizing the non-linearity. The modeling was carried out by ANN which consists of input, hidden and output layers. The input layer contained of the additives as effective variables while the output layer made of the calculated non-linear coefficients of 26 varistors. The structure of the hidden layer was determined by learning process to select the model with minimum root mean squared error (RMSE), percentage of absolute average deviation (AAD) and maximum coefficient of determination ($R^2$). The selected model was used to navigate the fabrication of ceramic including the effective narrow levels and the importance of the additives. The effective levels of the additives were TiO₂ (0.8–1.55), Bi₂O₃ (0.85–1.35), Mn₂O₃ (0.4–0.45), CoOₓ (0.4–0.8), Sb₂O₃ (0.4–0.5) and Al₂O₃ (0.00088–0.00138). The observed importance of the additives was TiO₂ > Bi₂O₃ > Mn₂O₃ > CoOₓ > Sb₂O₃ > Al₂O₃ in starting powder of the ceramic. Moreover, the predicted optimum point values of the additives that were experimentally verified. The result of the experimental verification was very close to the model prediction which confirmed the predictability of the model. As a conclusion, ANN has successfully modeled the initial additives in the starting powder of the ceramic core in the varistors. The model determined the optimum condition that maximized the non-linear property of the varistors. The higher quality varistor is able to protect the electronics from the overvoltage which enhance their lifetime. The longer lifetime instruments reduce vast amount of e-waste around the world.

Conflict of interest statement

The authors have declared no conflict of interest.

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