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Prediction of the Fracture Voltage of TiO_2-Doped ZnO–Bi_2O_3–MnO–CoO Ceramics Produced by the Chemical Precipitation Method with Using Artificial Neural Networks

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In this study, TiO_2 (2.5 and 5.0 wt%) doped ZnO–Bi_2O_3–MnO–CoO were produced by chemical precipitation method. In the ceramics produced, the effect of TiO_2 addition, sintering temperature, and time on breakdown voltage was experimentally measured and a mathematical model was developed according to these results. Based on the developed mathematical model and experimental results, an artificial neural network model was developed to determine the effect of TiO_2 on the breakdown voltage depending on the sintering temperature and time and was estimated by the breakdown voltage values. The results of the mathematical model and the artificial neural networks were statistically compared with the t test.

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1. Introduction

Electrical properties vary according to material groups. ZnO varistors are semiconductor ceramic materials. The electrical behaviours of multicomponent oxide ceramics depend on both the microstructure of the material and the products formed on the grain boundaries of ZnO [1]. ZnO varistors are ceramic devices with highly non-linear current–voltage characteristics similar to back-to-back Zener diodes. They are produced by sintering ZnO powder together with small amounts of other oxide additives [2, 3]. Almost no current flows under the voltage are known as the varistor voltage. When the voltage value exceeds the operating voltage, the resistance drops rapidly and begins to draw current. With varistors, it is usually intended to prevent the voltage from rising further by entering the circuit in the over-voltages, parallel to the entering of electronic systems [4].

Artificial neural networks (ANN), which cover studies based on computer learning, are a highly sought-after field of research under artificial intelligence. ANN technology is a new information processing approach based on the principle of imitating the behaviour of the human brain and nervous system in the computer world [5]. An ANN is a computational tool borrowed from the behaviour of biological neurons [6, 7]. Neural networks may estimate the output data based on presented input data (so called training sequence). Importantly, during the training process the network may gain the ability of predicting the output values without determining the dependence between it and the input values [8–10]. For more information on artificial neural networks, see [11–14].

The effect of TiO_2 on the microstructural and electrical properties of low voltage varistors has been observed in laboratory environments. Since the experiment is both time-consuming and costly, the breakdown voltage results for the effect of TiO_2 were produced by using ANN. ANN and test results were compared with t test.

In this study, a model developed for predicting the parameters used while determining the effect of TiO_2 on the electrical properties of low voltage varistors produced by chemical methods with artificial neural networks is explained.

Yüksel et al. (2003) describe the flow diagram of the study to observe the effect of TiO_2 addition on the electrical properties of the ZnO–Bi_2O_3–MnO–CoO varistor system produced by chemical methods.

The following parameters were used to determine the effect of TiO_2 on the electrical properties of low voltage varistors produced by the chemical method:

1. sintering temperature,
2. sintering time,
3. breakdown voltage.

Two different powder compositions were used in the study. The first composition DVB contained ZnO – 0.50 mol% Bi_2O_3 – 0.50 mol% MnO – 1 mol% CoO – 0.25 mol% TiO_2. The second composition contained

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ZnO – 0.50 mol% Bi_2O_3 – 0.50 mol% MnO – 1 mol% CoO – 0.50 mol% TiO_2 in DVC. The main variable here was TiO_2. The additive percentages were increased respectively and the effect values between them were determined. Experimental results of DVB and DVC compositions have been obtained in laboratory experiments depending on the sintering temperature and the duration of sintering.

Experiments were conducted in the laboratory to determine the effect of 0.25 mol% and 0.5 mol% TiO_2 on the microstructure and electrical properties of low voltage varistors produced by chemical methods. Sintering temperature and duration of sintering were determined as variable parameters in experiments. The sintering temperature was between 1150 and 1300 °C and the sintering time was between 1–2–3 and 6 h. Density, breakdown voltage, and grain size of the material were obtained in experiments performed at different sintering temperatures.

The breakdown voltage is V/mm, corresponding to 1 mA/cm^2. The mathematical model is determined as follows:

$$E_k = I_t + S_t T,$$

where E_k — breakdown voltage (1 mA/cm^2), T — temperature (°C), I_t — intersection (constant time), S_t — inclination (constant time).

Sixteen experimental data sets were obtained at the end of the experiments performed in the laboratory environment and the values obtained with the above mathematical model are given in Table I for DVB and in Table II for DVC.

TABLE I

As shown in Table I, the experimental breakdown voltage of DVB composition at 1150 °C for 1 h sintering was 80, and the calculated breakdown voltage value was measured as 80.

Sintering temp. [°C]	Sintering time [h]	Experimental results	Mathematical results
1150	1	80	80
1150	2	70	70
1150	3	60	60
1150	6	59	–
1200	1	66	66
1200	2	57	58
1200	3	48	50
1200	6	41	–
1250	1	50	52
1250	2	–	–
1250	3	50	40
1250	6	20	39
1300	1	40	35
1300	2	35	30
1300	3	30	–
1300	6	–	–

TABLE II

Results of the experimental and calculated values for the DVC (ZnO – 0.5% Bi_2O_3 – 0.5% MnO – 1% CoO – 0.50% TiO_2) and the results of the calculated values

Sintering temp. [°C]	Sintering time [h]	Experimental results	Mathematical results
1150	1	65	65
1150	2	58	58
1150	3	51	52
1150	6	44	–
1200	1	54	54
1200	2	48	50
1200	3	42	46
1200	6	40	–
1250	1	43	43
1250	2	38	41
1250	3	36	39
1250	6	30	–
1300	1	–	–
1300	2	33	33
1300	3	31	33
1300	6	28	–

As shown in Table I, the experimental breakdown voltage of DVB composition 1150 °C for 1 h sintering was 80; the calculated breakdown voltage value was measured as 80.

2. Experimental and mathematical measurement of breakdown voltage

Experiments were carried out in laboratory to determine the effect of TiO_2 on the electrical properties of low voltage varistors produced by chemical methods at 0.25 and 0.50 mol ratios. Sintering temperature and duration of sintering were determined as variable parameters in experiments. The sintering temperature was between 1150 and 1300 °C and the sintering time was between 1–2–3 and 6 h. The breakdown voltages of the materials produced at different sintering temperatures and sintering times were measured [15].

It is both time consuming and costly to calculate the breakdown voltage of the material by conducting experiments in the laboratory environment. For this reason, the breakdown voltage was calculated by increasing the number of data without performing an experiment. Sixteen data sets for breakdown voltage were produced by reducing the sintering temperature and the sintering time by changing the upper and lower limits of the sintering period. With the developed mathematical model, the breakdown voltage was calculated at different sintering temperature (1150–1300 °C) and at the sintering time (1–2–3–6 h). Possibility of a significant relationship between the data sets generated and the data sets obtained experimentally was statistically tested.

3. Recommended artificial neural network model

57 data sets obtained experimentally were used as training set in ANN and 57 data sets produced by mathematical model were used as test set in artificial neural networks. The validity of the mathematical model was also tested with ANN. The log-sig function is used as the activation function

$$f(x) = \frac{1}{1 + e^{-n}}.$$

A single layer model is used in ANN model. In the input layer, 4 process elements, 8 hidden layer process elements, and 1 output process element are used in the output layer. The activation function is between 0 and 1. However, the model results are in the range of −44.8 to 1300. For this reason, these results are normalized and reduced to the range of 0 to 1. The following formula has been used to normalize values:

$$V_N = 0.8\frac{V_R - V_{\min}}{V_{\max} - V_{\min}} + 0.1,$$

where V_R — data to be normalized, $V_{\min}$ — the data with the smallest value, $V_{\max}$ — the data with the largest value.

Figure 1 shows the comparison of the mathematical model results and the normalized values of the ANN results for the breakdown voltage values of the DVB composition. The ANN values of the breakdown voltage according to Fig. 1 approach 98% of the mathematical model. This situation supported the regression curve. In Fig. 1, only 16 of the 57 normalized data sets were used for experiments performed between 1150–1300 °C for 1–2–3 and 6 h.

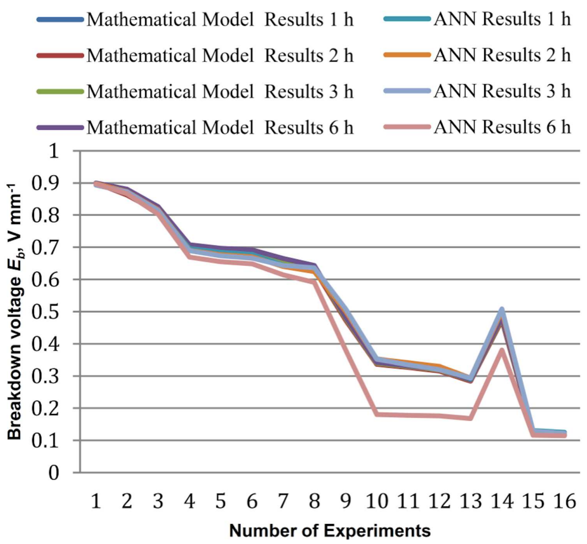

Fig. 1. Mathematical model for the DVB composition and the results of the breakdown voltage obtained by ANN.

Figure 2 shows the comparison of the mathematical model results and the normalized values of the ANN results for the breakdown voltage values of the DVC composition. The ANN values of the breakdown voltage according to Fig. 1 approach 98% of the mathematical model. This situation supported the regression curve. In Fig. 1, only 16 of the 57 normalized data sets were used for experiments performed between 1150–1300 °C for 1–2–3 and 6 h.

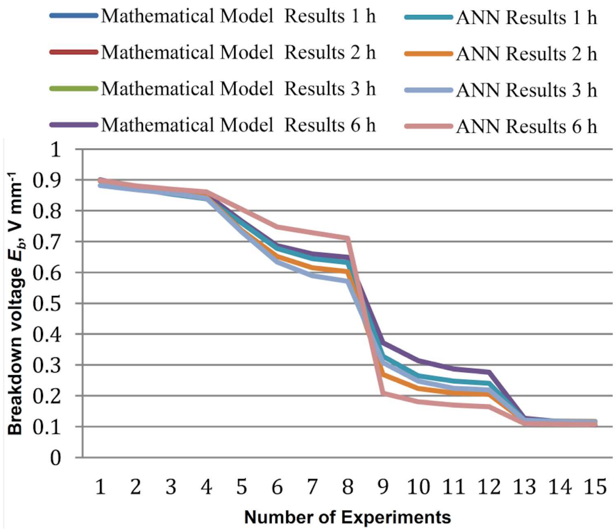

Fig. 2. Mathematical model for DVC composition and breakdown voltage results obtained by ANN.

4. Statistical analysis and results

The purpose of this test is to investigate whether the two models give approximately the same results as each other, that is, whether they have equal average. For this, t test was done in SPSS program. The hypothesis in this test is that there is no difference between the averages of the two models, that is, the averages are equal. We can show this with the H0 hypothesis — H0: $\mu_1 = \mu_2$; the alternative hypothesis is the H1 hypothesis, which indicates that the H0 hypothesis is rejected and that the averages are not equal. H1: $\mu_1 \neq \mu_2$, where red zone is defined as $z > Z_k$. According to $\alpha = 0.05$ level of significance is $Z_k = 1.96$.

As shown in Table III, the calculated z statistics for DVB and DVC combinations are smaller than Z_k. For each compound, the H0 hypothesis is accepted. That is, the mathematical model and the ANN model averages are not different from each other, the results produced by the two models are identical.

TABLE III

Z_k values for DVB and DVC

Sintering time	z test statistic		Z_k
	DVB	DVC	
1 h	0.213	0.179	1.96
2 h	0.167	1.044	
3 h	0.086	1.037	
6 h	1.572	0.577	

5. Conclusion

In this study, the parameters used in determining the effect of TiO_2 on the microstructural and electrical properties of the low voltage varistors produced by chemical methods were estimated by ANN. For this study, 16 experiments were carried out in the laboratory and results were obtained. However, the conducting experiments in the laboratory was both time consuming and costly. Therefore, ANN model has been developed in order to obtain fracture voltage, grain size and density. 57 educational sets and 57 test sets were used for estimation with ANN. In the study, it was observed that the ANN model has learned 98%. When sintering temperature, sintering time, intersection I_t, and slope S_t value are given with developed ANN model, break voltage, grain size, and density values can be estimated without experimenting in laboratory.

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