Relationship between Micro-parameters and Non-linearity of Zinc Oxide Varistor— A Computer Simulation Study

Cao Ze-Chun & Song Run-Sheng

Shanghai University of Science and Technology, Shanghai 201800, People's Republic of China (Received 14 September 1989; revised version received 2 January 1990; accepted 22 January 1990)

Abstract

An attempt is made to predict the non-linearity of a zinc oxide varistor (ZNR) by means of computeraided simulation. An equivalent circuit of the microunit (including grain and grain boundary) and a series-parallel equivalent circuit which is made from the micro-units instead of a random network are suggested, and the observed data of the micro-units are then fitted into equivalent circuits in sequence for computer simulation. The simulation results agree with the observation, and give an insight into the correlation between micro-parameters and nonlinearity of the ZNR.

Diese Arbeit beschäftigt sich mit der Vorhersage der Nichtlinearität von Zinkoxidvaristoren (ZNR) mit Hilfe der Computersimulation. Zur Simulation wird eine nicht zufällige, äquivalente Schaltung verwendet, die aus Mikroeinheiten (einschließlich Korn und Korngrenze) besteht. Diese äquivalente Schaltung wurde an die experimentellen Ergebnisse angepaßt. Die Simulationsergebnisse stimmen mit den Beobachtungen überein und geben einen Hinweis auf die Korrelation zwischen den Gefügeparametern und der Nichtlinearität von ZNR.

On tente de prédire la non-linéarité de varistances d'oxyde de zinc (ZNR) à l'aide d'une simulation assistée par ordinateur. La microstructure est assimilée à un circuit équivalent série-parallèle non aléatoire constitué d'unités (grain et joint de grains). Les données expérimentales sur les unités sont disposées en circuits équivalents pour la simulation par ordinateur. Les résultats de la simulation sont en accord avec l'observation et donnent une idée de la corrélation entre les paramètres de la microstructure et la non-linéarité des ZNR.

1 Introduction

It is generally accepted that the outstanding I-V characteristic of a zinc oxide non-linear resistor (ZNR) is related to the grain boundaries (GB) distributed throughout the ZNR ceramic body. The non-linear coefficient, α , is taken as the most important performance parameter for a ZNR component, but an apparent discrepancy has been noted for a long time: the α -value is found to be greater for a GB (ranging from 60 to 100) than for the ceramic body ($\alpha \simeq 40$).

In the past two decades, a number of papers have related the α -value of a ZNR to its composition and processing; all theories are based on non-linear behavior of a single GB. Therefore there is a gap existing between the microscopic parameters and the non-linearity of the ceramic body. An attempt has been made to establish a bridge across this gap; for example, Emtage suggested a statistical approach,¹ but he could not explain why the α -value of a ceramic body is generally less in a ceramic than in an isolated GB.

In fact, the actual macroscopic component of ceramics involves a random network of single GBs, the performances of which are more or less different, and little or no progress has yet been made on the macroscopic system of a random network.²

In our view, for this large number of GBs, the use of statistics is necessary, fitted with a network model. A similar computer-aided simulation study of a ZNR component supports the suggestion;³ the nonlinear coefficient of a ZNR lightning arrester component was simulated by use of the α -values of thin wafers cut from the component, with a seriesparallel equivalent circuit instead of a network. The simulation result is in good agreement with the experimentally determined α -value, hence in this

85

paper the approach is adopted to simulate the nonlinearity of a small macroscopic region (SMR) by the microscopic parameters.

2 GB Model and Micro I-V Characteristic Curve

2.1 GB model

According to current theory, the distinctive feature of the ZNR GB is its double Schottky barrier. However, further research shows that part of the GB is dominated by a Bi₂O₃ segregation layer with its α -value ranging between one and two. In addition, according to our observation and Schwing and Hoffmann's,⁴ there may exist another kind of GB which has neither Bi₂O₃ segregation nor a distinct barrier effect, and its resistivity is comparable with that of the grain. In the above three kinds of GBs, the first one is the source that leads to the non-linear character, so it may be called the 'effective GB'. The percentage of effective GBs can be estimated as follows. In an experimental observation the thickness of wafers cut from a ZNR component is 0.94 mm and the average grain size in the ceramic is ~ 10 μ m; hence there are ~ 100 grains along the direction of wafer thickness. If there were Schottky barriers in every GB, the breakdown voltage of the wafer should be ~ 340 V, but the observed value is only 140-150 V, therefore the percentage of effective GBs may be 40%.

The equivalent circuit of the grain (including GB) is shown in Fig. 1. In the figure, besides the resistor which represents the grain, there are three kinds of element which express the different GB characters; these emerge randomly and exclude each other.

2.2 Microscopic I-V characteristic

According to the experimental data, I-V curves of the effective GBs consist of two regions. Both vary in exponential fashion, but their values of power factor, α , are different. Thus the I-V characteristic can be formulated as follows:

$$I = V^{\alpha_1}/C \qquad V < V_{\rm b} \\ I = (V - V_1)^{\alpha_2}/C \qquad V \ge V_{\rm b}$$
(1)

where V_b is the breakdown voltage and $V_1 = V_b - (V_b^{\alpha_1})^{-\alpha_2}$. This definition of V_1 ensures that the two parts of the *I*-*V* curves link up at the breakdown point, $\alpha_2 \simeq 100$, $\alpha_1 < 5$. *C* is a constant derived from the experimental *I*-*V* curve.⁵ There are seven sets of data available. The *I*-*V* characteristic of the Bi₂O₃ segregation layer is also formulated from the same source:⁵

$$I = V^{1 \cdot 39} / e^{14 \cdot 13} \tag{2}$$



Fig. 1. The equivalent circuit of grain.

3 Influence of micro-parameters on non-linearity

In this section, when the α -values of SMR are simulated, the following expression is used:

$$\alpha = \log(I_2/I_1) / \log(V_2/V_1)$$
(3)

The I and V data fitted into the expression are obtained from the simulation. The current, I, ranges from 10^{-6} to 10^{-3} A and the corresponding voltage, V, is ~130–170 V.

3.1 Effect of GB breakdown voltage distribution

It is accepted that the breakdown voltage of a GB Schottky barrier is 3.4 V, but in fact the breakdown voltage has its own distribution.

To study the influence of the voltage distribution on the α -value, four distribution characters are chosen. The first three are for the assumed case and the fourth is an experimental observation,⁵ and the various distribution characters are illustrated in Fig. 2. It can be seen from Fig. 2 that case A is an ideal and simplified condition, as 100% GBs have the same breakdown voltage, 3.4 V. The standard deviations of cases B and C are 0.003 22 and 0.010 58, respectively, but the mean values are the same, i.e. 3.4 V. For case D, the mean value is 3.5 V and its standard deviation is 0.0193. Table 1 shows that the α -value becomes larger as the data of breakdown



Fig. 2. Various distributions characters of breakdown voltage.

Table 1. Influence of the distribution of breakdown voltage

Distribution of breakdown voltage	В	С	D	
α-Value	79.2	46.5	40.5	38.3

voltage are more concentric. The above result is obtained when bulk resistivity of the grain is taken as 0.1Ω -cm,⁶ and the ratio of effective GB is 40%. In the simulations below, unless otherwise stated, the following micro-parameters are adopted: the ratio of effective GB is 40%, grain resistivity is 0.1Ω -cm and the distribution of GB breakdown voltage is case D.

3.2 Effect of bulk resistivity and ratio of Bi_2O_3 segregation

In Table 2 the influence of bulk resistivity on the α -value is shown; this influence explains the choice of 0.1 Ω -cm resistivity.

Table 2. Influence of bulk resistivity of grain

Bulk resistivity (Ω-cm)	0.1	0.01	0.001	0	
α-Value	38.3	105	129	247	

Table 3. Effect of Bi₂O₃ segregation layer

$\begin{array}{c} Bi_2O_3\\(\%)\end{array}$	0	10	20	30	x-Value observed	
α-Value	38.3	29.8	26.2	24.0	30	

In Table 3 a further parameter, the Bi_2O_3 segregation layer (i.e. the percentage of GBs for which their properties are dominated by the Bi_2O_3 segregation layer), is added, and its influence on the α -value is shown and compared with the observed α -value.

3.3 Effect of distribution of effective GBs

Because the fluctuation of local chemical composition and structure is unavoidable, one may expect that the distribution of effective GBs is different in each micro-region. Therefore it may be considered that in the equivalent circuit shown in Fig. 1 the number of effective GBs in each branch is different, and this difference in distribution should affect the α -value of the SMR. In Table 4 three kinds of distribution are given, and the α -value for each distribution is simulated.

Table 4. Three kinds of distribution of effective GBs

Effective GB — (%)		Branch number								α-Value	
	1	2	3	4	5	6	7	8	9	10	
Case A	40	40	40	40	40	40	40	40	40	40	26.3
Case B	36	38	38	40	40	40	40	42	42	44	17.7
Case C	30	32	35	36	41	42	43	46	47	48	14.9

4 Discussion and Conclusion

In this paper the micro-parameters involved are ratio of effective GBs and its distribution, distribution of breakdown voltage, bulk resistivity, and percentage of Bi_2O_3 segregation layer in GB. All of these parameters have an effect on the non-linearity of the SMR. Of these parameters, only two, i.e. ratio of Bi_2O_3 segregation and bulk resistivity, are controlled directly by chemical composition; the others are related to non-homogeneity. This accounts for the necessity for strict control of processing.

With respect to the role of these parameters, it is noted that the percentage of the Bi_2O_3 layer has less effect on the α -value. This simulated result does not coincide with the observation, as it is recognized that the Bi_2O_3 layer is harmful to non-linearity.⁷ This result may have arisen from eqn (2), according to which the Bi_2O_3 layer has rather high conductivity. On the other hand, distribution of effective GBs is worth attention, as it has a profound effect on the non-linearity.

In a word, the simulation gives us plentiful information about the correlation between microparameters and non-linearity, although this information is mainly qualitative rather than quantitative. For a more complete and quantitative simulation, further accumulation of experimental data for the ZNR is needed.

This paper suggests an approach to correlate micro-parameters with macro-performance, not only for a ZNR but also for other kinds of ceramics.

References

- 1. Emtage, P. R., Statistics and grain size in zine oxide. J. Appl. Phys., 50 (1979) 6833-7.
- Mahan, G. D., Theory of ZnO varistors. In *Grain Boundaries* in *Semiconductor*, ed. H. J. Leamy. Elsevier Applied Science, New York, 1982, pp. 333–41.
- Song, R. S. & Cao, Z. C., A computer-aided simulation for the nonlinearity of zinc oxide varistor (in Chinese). J. Shanghai Univ. Sci. Technol., 12(2) (1989) 44–51.

- Schwing, U. & Hoffmann, B., New approach to the measurement of the single-contact varistor. In Grain Boundary Phenomena in Electronic Ceramics, ed. L. M. Levinson. American Ceramic Society, Columbus, OH, 1981, pp. 383-93.
- 5. Einzinger, R., Grain boundary properties in ZnO varistor. In Grain Boundary Phenomena in Electronic Ceramics, ed. L. M.

Levinson. American Ceramic Society, Columbus, OH, 1981, pp. 359-74.

- 6. Einzinger, R., Grain junction properties of ZnO varistor. Appl. Surface Sci., 3 (1979) 390-408.
- 7. Takemura, T., Kobayashi, M., Takada, Y. & Sato, K., Effects of bismuth sesquioxide on the characteristics of ZnO varistors. J. Am. Ceram. Soc., 69 (1986) 430-6.