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Statistical Study of the Thin Metal-Oxide Varistor Ceramics

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Abstract: In this paper, computational methods are used to study statistically the conduction phenomena in the thin zinc-oxide non-linear varistors, used to protect the electrical and electronic circuits against the overvoltages. Firstly a Monte Carlo method is utilized to model the conduction phenomena in the varistors. This approach shows that the number of ZnO grains, providing the current path between the electrodes of thin varistors fits a lognormal distribution. Furthermore it is found that the "turn on" characteristics, conduction threshold voltage and the nonlinearity coefficient of these varistors can be controlled by the fraction of non-conducting grains.

Key words: Metal-oxide non-linear ceramics, ZnO varistors, Statistical model, Monte Carlo method

INTRODUCTION

A varistor is a type of resistor with a significantly non-ohmic current-voltage characteristic. The name is a portmanteau of variable resistor, which is misleading since it is not continuously user-variable like a potentiometer or rheostat and is capacitor rather than resistor at low field. Its most famous type is the metal oxide varistor (MOV), which is also called as ZnO varistor (Meshkatoddini, M.R., S. Boggs, 2006; Andoh, H., *et al*, 2000; Greuter, F. and G. Blatter, 1990; Shengtao Li, Jianying Li, 2002; Tao, M., Bui Ai, 1987; Shengtao, Li, Feng Xie, 1998; Robert, C.P. and G. Casella, 2004). These varistors are used to protect circuits against excessive voltages. They have become more and more important during the past three decades due to their highly non-linear electrical characteristics and their large energy absorption capacity. They are normally connected in parallel with an electric device to protect it against the overvoltages. They contain a mass of zinc oxide grains in a matrix of other metal oxides sandwiched between two plasma sprayed metal electrodes. The ZnO grains having dimensions in the range of 10µm to 100µm. The boundaries between the grains form double potential barriers with Schottky junctions with conduction voltages in the range of 3 .5V. The boundary between each grain and its neighbor forms a Zener-like diode junction. ZnO grains are separated by these "active" grain boundaries of nanometers thickness. Then the mass of randomly oriented grains is electrically equivalent to a network of back-to-back diode pairs, each pair in parallel with many other pairs. When a small or moderate voltage is applied across the electrodes, a small thermally activated reverse leakage current flows through the diode junctions. When a large voltage is applied, the diode junctions break down from the avalanche effect, and large current flows. The result of this behavior is a highly nonlinear current-voltage characteristic, in which the MOV has a high resistance at low voltages and a low resistance at high voltages (Fig. 1).

In this work we try to have a statistical study on these varistors, to find a suitable way to control their main characteristics such as the nonlinearity coefficient and conduction threshold voltage.

As seen in Fig. 1, three regions can be distinguished in the current voltage characteristics of the ZnO varistor. At low voltages, the insulating barriers between the grains result in a very high and almost Ohmic resistivity, which is called the pre-breakdown or Ohmic region. At a certain voltage, called the threshold or breakdown voltage, the system enters the breakdown region in which the current increases abruptly, and the dependence of current on voltage is described by the empirical relation:

$$I = k\ V^{\alpha} \qquad (1)$$

From which the parameter α is equal to:

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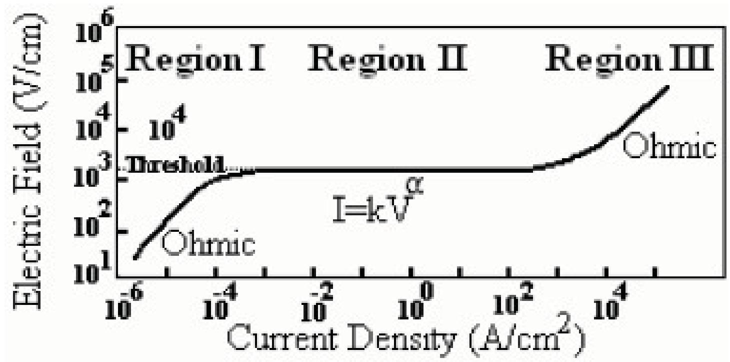

Fig. 1: Typical E-J diagram of a ZnO varistor

α = d [log *(I)]* / d [log *(V)]* (2)

This parameter is a measure of the element nonlinearity, which varies with voltage. At higher current densities, the voltage starts to increase again resulting in an upturn region of the I-V characteristic. This voltage increase gradually becomes linear with current, i.e. Ohmic, and is associated with the resistivity of the ZnO grains, i.e. the voltage drop in the ZnO grains.

In ZnO varistors, when a voltage is applied between the electrodes, a fraction of the grains contain no conducting boundaries with other grains, which results an arbitrary path for current across the ZnO element, being the number of crossed grains a statistical parameter. As well there is certain number of grains, which does not present any non-linear characteristic.

Few works can be found, which have experimentally studied the individual grain boundaries in the varistor (Tao, M., Bui Ai, 1987).

Most of the Schottky junctions give a nonlinearity coefficient equal to equation (2) which is normally in the range of 30-70 for a normal varistor, whereas the actual α of typical good ZnO material can be in the range of 150 and more. It can attain values greater than 200 in certain grain to grain microvaristors. Fig. 2 shows the typical variation of the current density as a function of the barrier voltage, for a single barrier in a typical varistor.

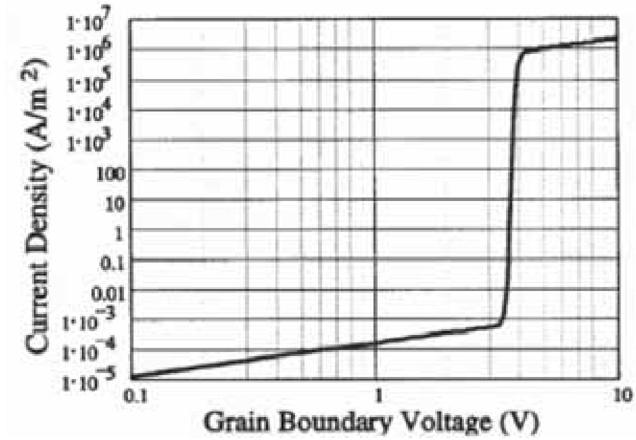

Fig. 2: The grain boundary current density vs. grain boundary voltage.

In Fig. 3 the variation of the non-linearity coefficient α as a function of the varistor barrier voltage, for a single potential barrier is observed. This curve is deduced computationally, using Maple software, from the slope of the current-voltage characteristic of a single grain boundary as in Fig. 2.

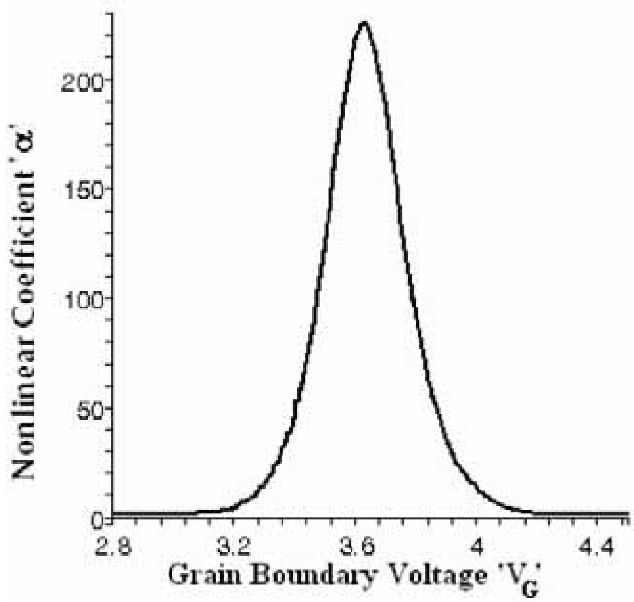

Fig. 3: Non-linearity coefficient α as a function of the varistor barrier voltage, for a single potential barrier (Meshkatoddini, M.R., S. Boggs, 2006).

MATERIALS AND METHODS

Varistor Microstructure:

Now we consider the typical microstructure of a ZnO varistor (Fig. 4).

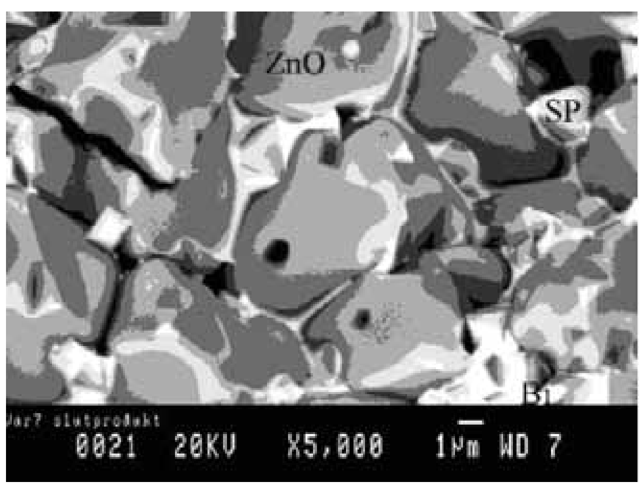

Fig. 4: Photo of the typical microstructure of a ZnO varistor. (ZnO=Zinc oxide grain, Bi=Bismuth, Sp=Spinel phase)

In Fig. 5 a simplified model of the varistor microstructure is observed. We use this model for computer simulation.

If the ZnO element thickness is D and the average grain thickness is d, then the minimum number of grain boundaries between the electrodes is L=D/d.

Monte Carlo Method:

Monte Carlo is a technique that provides approximate solutions to problems expressed mathematically

(Robert, C.P. and G. Casella, 2004; Ueberhuber, C.W., 1997). Using random numbers and trial and error, it repeatedly calculates the equations to arrive at a solution. Then using random numbers or more often pseudo-random numbers, as opposed to deterministic algorithms, uses this algorithm for solving various kinds of computational problems.

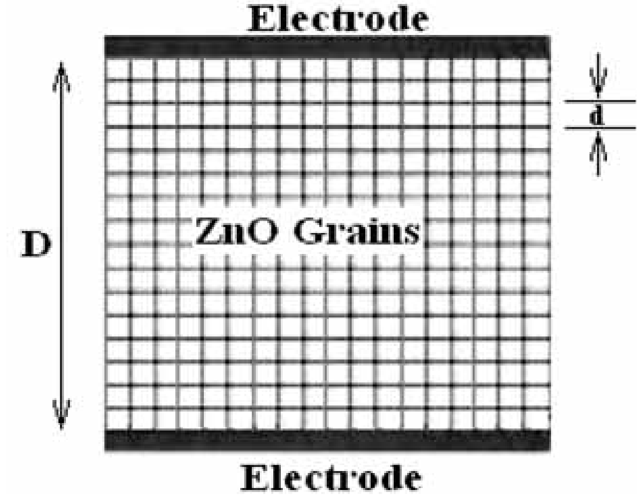

Fig. 5: Simplified micro-structural model of varistor for computer simulation.

Monte Carlo methods are extremely important in computational physics and related applied fields. Interestingly, the Monte Carlo method does not require truly random numbers to be useful. Much of the most useful techniques use deterministic, pseudo-random sequences, making it easy to test and re-run simulations. The only quality usually necessary to make good simulations is for the pseudo-random sequence to appear "random enough" in a certain sense. That is that they must either be uniformly distributed or follow another desired distribution when a large enough number of elements of the sequence are considered. Because of the repetition of algorithms and the large number of calculations involved, Monte Carlo is a method suited to calculation using a computer, utilizing many techniques of computer simulation.

Using a Monte Carlo algorithm, we followed a stochastic procedure to compute the number of the conducting grains on the current path in the varistor model as a statistical parameter. The flowchart of the used program is observed in Fig. 6. In this diagram the letters K and N denote, respectively, the iteration number and the variable for the number of each layer in micro-structural model of the varistor. B is the number of active grain boundaries through which the current passes in going from one electrode to the other. As well, we define the probability of a non-conducting grain boundary as P. For P⁼0, all grain boundaries are always active. It is obvious that the existence of non-conducting grains results in a longer path for current across the ZnO element, which depends on the fraction of non-conducting grains. We undertake a statistical analysis of the effect of L (the number of ZnO grain layers across the varistor) and P (the probability of a non-conducting grain boundary) on the nonlinear characteristics of the varistor as characterized by α.

As said above, for P=0, there is no non-conducting grains and all path lengths are the same, equal to L. With increasing fraction of non-conducting grain boundaries P, the conducting grains number B, augments substantially, which will increase the voltage per unit thickness of the ZnO element. P can also be augmented by increasing the amount of non-conducting inter-grain material, often as a by-product of attempting to reduce grain size. This non-conducting phase can be a spinel phase (Fig. 4). Obviously increasing the number of non-conducting grain boundaries increases the current density in the remaining grain boundaries and results in greater grain boundary power dissipation and temperature rise.

Numerical Computations:

By running the Monte Carlo program with different values of L, the number of ZnO grain layers across the varistor, and P, the probability of non-conducting grain boundaries in varistor, we obtained statistical sets of data for B, i.e. the number of grains crossed by the current.

As an example, a probability density histogram of B's data for the case of a very thin varistor with L=10 and P=0.3 is seen in Fig. 7, which is related to a varistor of about 0.1 mm thick.

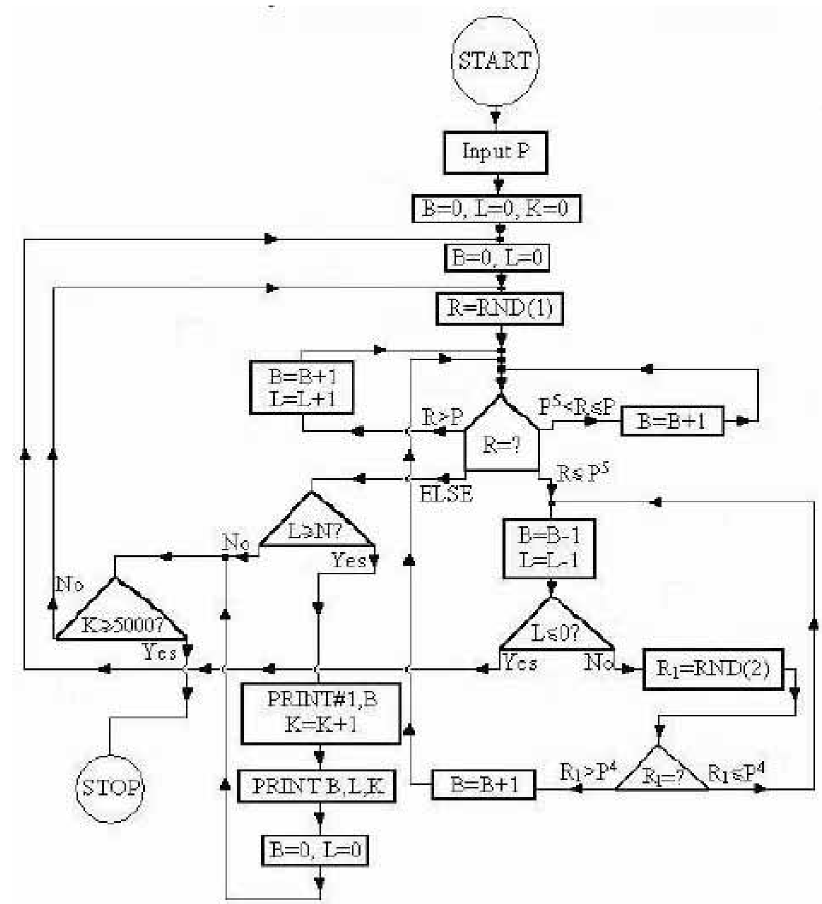

Fig. 6: Flowchart of Monte Carlo algorithm, used in this work, for computation of the number of grains on the current path through the varistor.

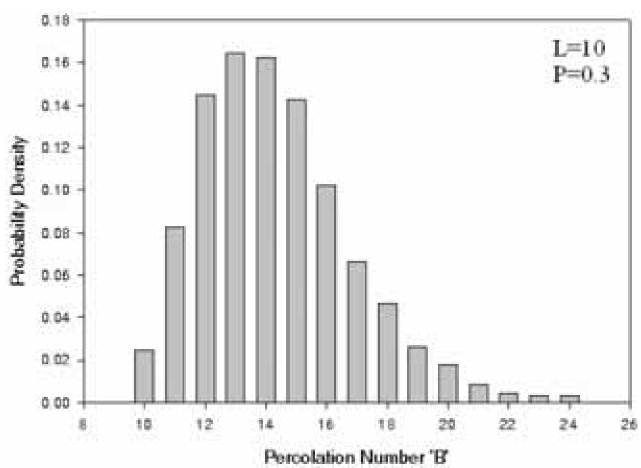

Fig. 7: A probability density histogram of the number of grains crossed by the current, obtained for special case of a very thin varistor of about 0.1 mm thick with probability of non-conducting grains equal to 30%.

We analyzed the statistical distribution of B by fitting different distribution curves on it. Several distributions such as Normal, Lognormal, Weibull, Logistic, Loglogistic and Exponential were used for fitting our computational data. The best fitness was seen to be for the three distributions of Normal, Weibull and Lognormal (identically for LogeNormal and Log10Normal), comparing to the others.

In Fig. 8 we can observe the fitted curves for these three distributions concerning the special case of Fig. 7.

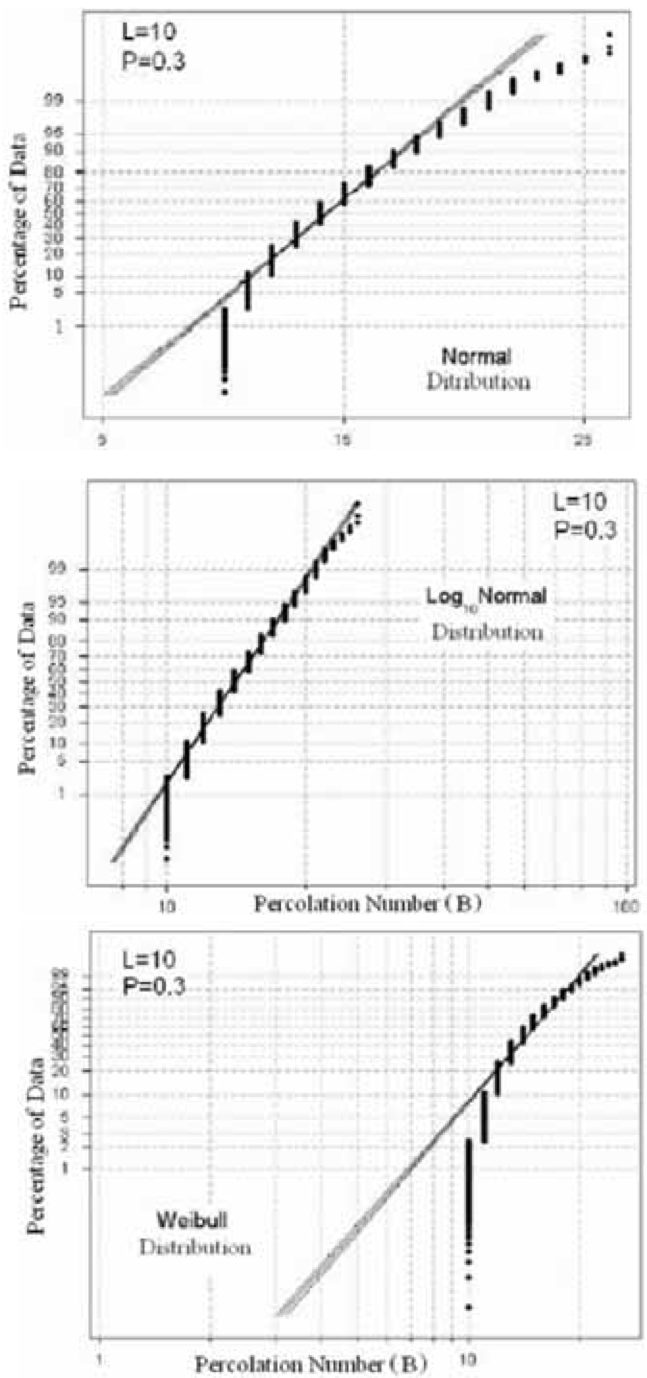

Fig. 8: Curve fitting of the number of grains crossed by the current, on three different distributions for the data of Fig. 6.

The Anderson-Darling statistic is a measure of how far the plot points fall from the fitted line in a probability plot. Using the Anderson-Darling measure to calculate the fit goodness of these distributions, we obtain the curves of Fig. 9.

The statistic is a weighted squared distance from the plot points to the fitted line with larger weights in the tails of the distribution. In this method, a smaller Anderson-Darling (AD) measure indicates that the distribution fits the data better.

As can be observed in Fig. 9, the LogNormal distribution has the best fit for the B data concerning the thin varistors of this study.

If the probability of a grain boundary to be non-conducting is P and as we supposed in our model that the grains are cubes, then it can be shown that to a first approximation, the mean number of active grain boundaries through which the current passes between electrodes is:

$$B = L\left(1 + \frac{P}{1-P}\right) \quad (3)$$

$$0 < P < 1$$

And as we realized in this work that the percolation number data for thin varistors obey the lognormal distribution, we write, using the Maple software, the relation (4) as an analytical formula for the standard deviation, s, of thin varistors data, having the lognormal distribution.

$$s = \sqrt{(e)^{\left(\frac{\left(\ln\left(L\left(1-\frac{P}{P-1}\right)\right) - \frac{\ln\left(L\left(1-\frac{P}{P-1}\right)\right)}{L}\right)^{2}}{L}\right)}\left((e)^{\left(\frac{\left(\ln\left(L\left(1-\frac{P}{P-1}\right)\right) - \frac{\ln\left(L\left(1-\frac{P}{P-1}\right)\right)}{L}\right)^{2}}{L}\right)} - 1\right)} \quad (4)$$

By plotting this equation for different values of L and P in Maple software, we obtain Fig. 10. As it is seen, the standard deviation is not high for amounts of P less than 0.5, while it is great for bigger P's in thinner varistor blocks.

RESULTS AND DISCUSSIONS

It was assumed in this work that each current path is independent of every other path. In fact, at large P, the number of non-conducting grain boundaries would reduce the likelihood of interconnection of paths. But in P less than 0.5, especially in thin varistors even if two paths of differing length are near each other, the probability of their having substantially differing potentials is not great.

We realize from the form of the statistical distribution for B, that as P increases, the varistor conduction turn-on will be more rapid. This can be deduced from the low B tail of the statistical distribution. For small L, the average value of B increases with P, but the minimum value of B, which is L, remains the same. Thus the ratio of the mean to minimum possible value of B increases. The turn-on characteristics are determined mainly by the first few paths to conduct. Thus the number or fraction of completed paths for various P must be considered in addition to α.

Fig. 11 compares lognormal and Normal distributions with the same mean (200) and with variances selected to give the same minimum value (~100) in a population of 600 random numbers. This figure indicates that the probability density of B increases much more rapidly at low values of B for the lognormal than for the Normal distribution. Thus conditions, which drive the statistical distribution for B toward the lognormal distribution, are likely to result in more rapid turn-on of the varistor element. The lognormal distribution also has a long tail at high values, which will cause a long tail in α. Based on the numerical computations and the distributions thereof, we believe that the more rapid turn-on as a function of increased P for large L (thick elements) is probably associated with a transition from a Normal distribution at P=0 toward a lognormal distribution with increasing P.

This transition can be rationalized from the probability density of B for a thin varistor with a reasonable probability of non-conducting grains, for which the distribution is clearly asymmetric with a rapid turn-on, when the shortest path across the arrester becomes conducting, followed by a rapid increase in the number of conducting paths with increasing voltage.

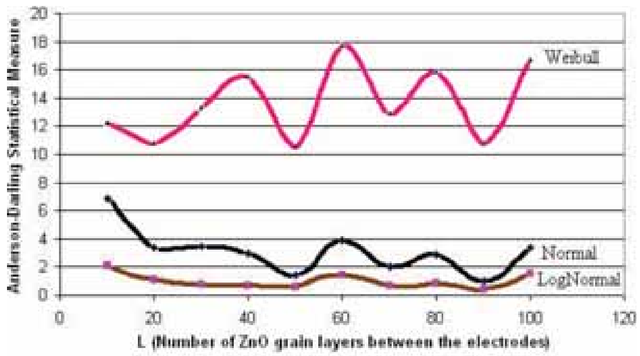

Fig. 9: Comparing the fitness of three different distributions on our data, concerning varistors of 0.1 to 1 mm thick.

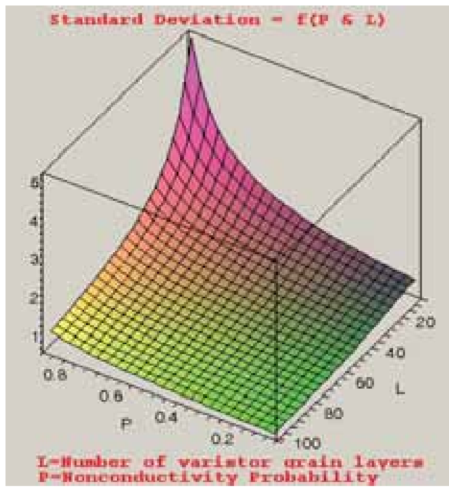

Fig. 10: Standard Deviation for percolation number B of thin varistors, having lognormal distribution.

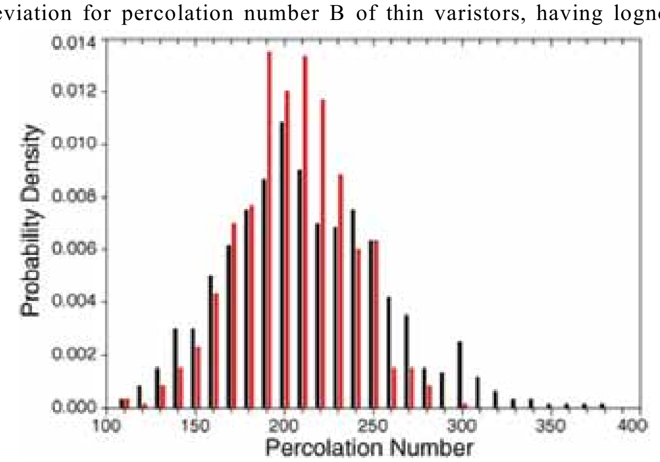

Fig. 11: Comparison of Lognormal (black) and Normal distribution data with same mean (200) and with variances set to give about the same minimum value (100) in a population of 600.

In Fig. 12, the variation of α as a function of the applied voltage is seen for a non-conducting grain probability 0.5 for a thick varistor of 10 mm thickness (L=1000) and a thin varistor of 0.5 mm (L=50) thickness.

Both cases result in asymmetrical α characteristics, while the varistor thickness has an obvious influence on the shape of the curve. We accomplished the same analysis for varistors with different thickness (Number of grain layers L) and probability of non-conducting grains (P).

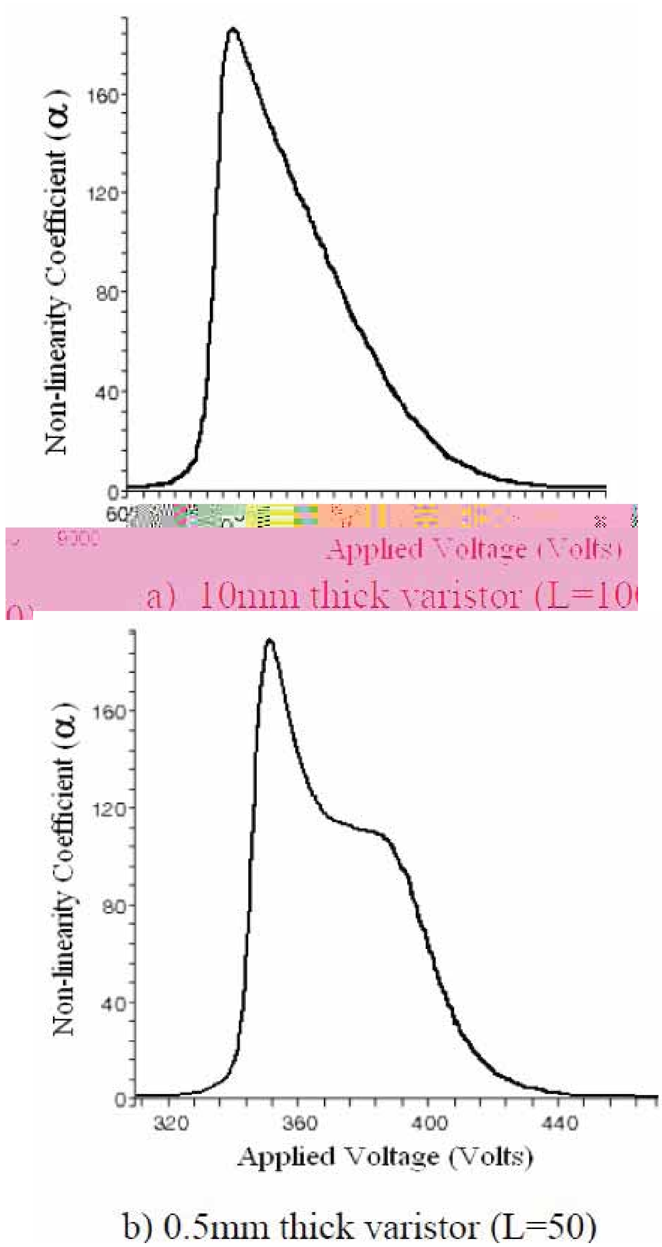

Fig. 12: Varistor nonlinearity coefficient α as a function of the applied voltage for thin and thick varistors at a non-conducting grain probability of 0.5.

To provide a basis for comparison of the α(V) curves, we define the parameters FWHH and β as measures for broadness and rate of rise of the α(V) curve (Fig. 13). FWHH is the Full Width at Half Height of the curve and β is defined as:

$$\beta = (V_{90\%} - V_{10\%}) / V_{10\%} \quad (5)$$

According to this definition for β, the small β means large slope of α(V) curve.

In Fig. 14, we see the variation of the (FWHH) of the α(V) curves as a function of L, which is linear as might be expected.

For P=0 and large L, the characteristics are just a multiple of the grain boundary characteristics which are modeled as symmetric. As the fraction of non-conducting grains increases, the mean percolation path increases and the probability of a short percolation path decreases.

However the minimum possible path remains the same (L) and above the minimum path, the number of conducting paths appears to increase rapidly which results in an asymmetric α with more rapid turn-on.

As both the threshold voltage and width of α are the sum of the contribution from each grain, i.e. a 10 mm thick varistor (L=1000) is equal to twenty 0.5 mm thick (L=50) varistors in series, so that the I(V) curve of the former will be sum of the I(V) curves of the latter. Thus FWHH(L) should increase linearly with L, and the result of Fig. 14 can be taken some degree of verification of the computational methods.

Fig. 15 shows the variation of β with non-conducting grain probability (P) for a thin (L⁼50) and a thick (L=1000) ZnO element. As we can see in this figure, the rate of rise of the α(V) curve depends on both the non-conducting grain probability and the element thickness.

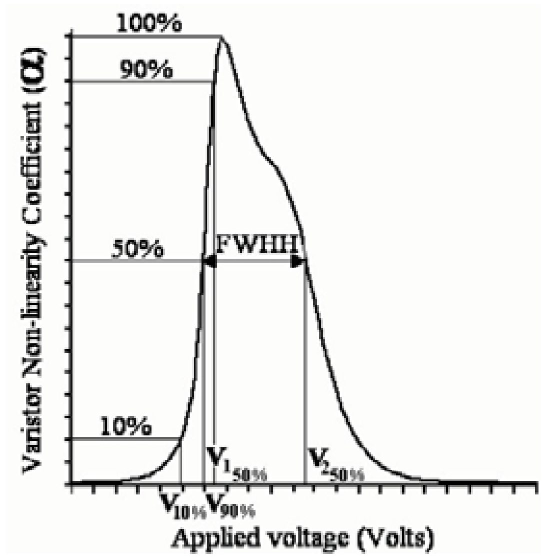

Fig. 13: Definition of parameters FWHH and β.

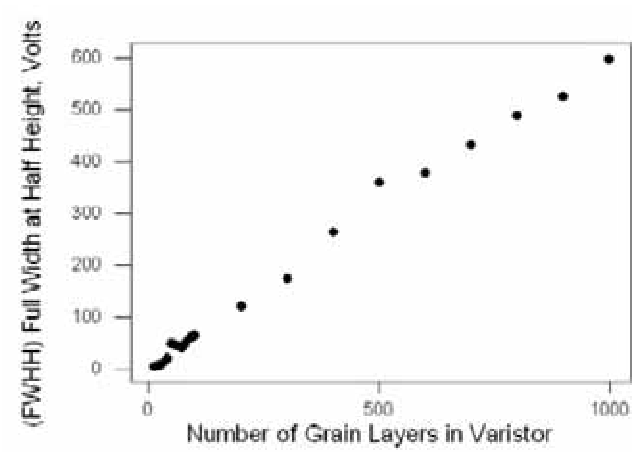

Fig. 14: Full Width at Half Height (FWHH) of the α(*V*) curves with P=0.5 as a function of L.

For P>0.5 and large L (thick varistor), β remains constant, which means that increasing P has little effect on the varistor turn-on characteristics. This is probably a result of the fact that for large L, the standard deviation in B decreases as a fraction of L, so that the extreme value in B decreases relative to the mean.

With increasing P, the mean number of grain boundaries increases, as does the distribution of the number of grain boundaries through which the current passes from one electrode to the other. As the grain boundary characteristic is highly nonlinear, the conductivity of the ZnO element rises rapidly once the first few current paths become conductive. This probably accounts for the increasing asymmetry in α for the whole varistor, and increasingly rapid current onset with increasing P, as the statistical distribution of path lengths broadens with P.

For large L, β decreases with increasing conducting grain probability, P, but for small L this is not the case. As well, the peak value of α increases with increased P for thick elements but not for thin elements. This must result from competition between the larger variance in B, the percolation path for small L, with the nature of the tail of the statistical distribution at low values of B, which determines the turn-on characteristics.

One would prefer the ZnO "turn on" (become substantially conductive) to be very rapidly so that the AC operating voltage can approach more closely the protection level of the varistor without causing excessive power dissipation. On the other hand, how the varistor approaches its ultimate conductivity with voltage is less important. As we observed, the probability, P, of non-conducting varistor grains has an influence on parameters such as the rate of rise of α(V) curve.

Conclusion:

In this work the characteristics of the thin ZnO varistors were statistically studied. The number of ZnO grains on each conducting path through a ZnO varistor, crossing by the current, is a statistical parameter (B).

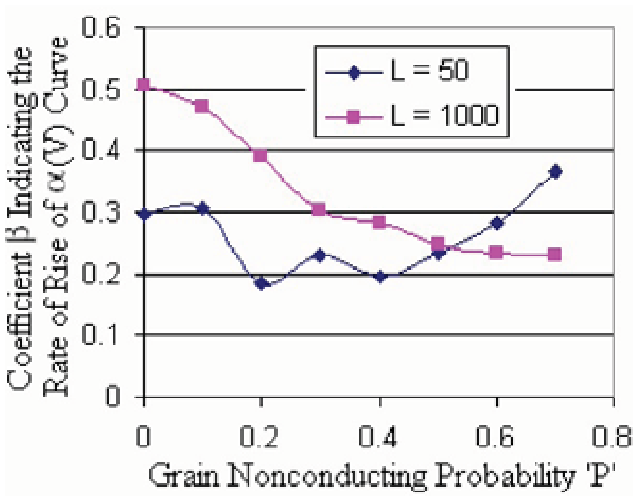

Fig. 15: Variation of β as a function of P, for L=50 (0.5 mm thick) and L=1000 (10 mm thick) varistors. Smaller β tends to indicate more rapid "turn-on" of the varistor with applied voltage.

The effect of the fraction of non-conducting grains as a function of the element thickness was studied to see how its properties, such as B, change as the number of grains across the varistor drops from 1000. It was seen that the distribution of B depends on the block thickness and percentage of non-conducting grains in the varistor.

A Monte Carlo method was used for investigation of the statistical behaviour of the varistors of 0.1 to 5 mm thick. Using this algorithm, it was found that the number of ZnO grains providing the percolation path in thin varistors, up to 1mm thick with P smaller than 0.5, follows a lognormal distribution.

It was shown that the nonlinearity of ZnO ceramics can be controlled, to some degree, by the fraction of non-conducting grains. Thus we can choose the best value for P to have the maximum rate of rise of the α(V) curve. This will result in a rapid "turn on" of the ZnO element, which allows the circuit being protected to operate more closely to the protection level without excessive power dissipation in the arrester element. This optimum value of P certainly depends L, which is related to the thickness of the varistor.

With increasing fraction of non-conducting grain boundaries P, the percolation number B, increases substantially, which will increase the voltage per unit thickness of the ZnO element. This can be exploited commercially in order to increase the percolation number.

The results of this work can help us to have a better understanding of the behavior of these varistors, and the dependence of this behavior on their geometrical dimensions and the constituting materials. This will also enable us to have more realistic electric models for these ceramic elements.

Computer Programs:

Basic Program for Monte Carlo Modeling:

```
4 OPEN "D:\Mohammad\B data\L200_1000. K5000. P 0.5\L200_K5000_P0.5.DAT" FOR APPEND AS #1
'P = Probability of nonconducting grain
5 P = 0.5
'LMAX = Number of the layers across the varistor
6 LMAX = 200
'KMAX = Number of iterations (Current injection to the upper electrode)
7 KMAX = 5000
10 B = 0 : L = 0: K = 0
15 B = 0 : L = 0
20 R = RND(1)
30 IF R > P THEN GOSUB 100
40 IF P >= R AND R > P^5 THEN GOSUB 200
50 IF R <= P^5 THEN GOSUB 300
60 IF L >= LMAX THEN GOSUB 400
70 IF K >= KMAX THEN GOTO 450
80 GOTO 20 100 B = B + 1 110 L = L + 1 120 RETURN 200 B = B + 1 210 RETURN 300 B = B - 1
310 L = L - 1
320 IF L <= 0 THEN GOTO 15
330 R1 = RND(2)
340 IF R1 > P ^ 4 THEN B = B + 1
350 IF R1 <= P ^ 4 THEN GOTO 300
360 RETURN 400 PRINT #1, B 410 K = K + 1
420 PRINT B, L, K
430 B = 0: L = 0 440 RETURN 450 STOP
500 END
```

Maple Program for Varistor's Conduction Modeling

```
restart;
B:=L*(1+sum(P^n, n=1..infinity)); # B=The mean
number ofactive grain boundaries through which the current passesbetween electrodes. s1:=(log(B) - (1/L)*(log(B))) / (sqrt(L));
s := (((exp(1))^(s1^2))*(((exp(1))^(s1^2))-1))^(1/2); readlib(log10);
plot3d(s,P=0.1. .0.9, L= 10. .1000,axes=boxed, title="Standard Deviation = f(P & L), P=Nonconductivity Probability, L=Number of varistor grain layers");
J:=10^((tanh(50*log10(VG)-28))*4.5-
5.5) *10^( (500*log10(VG) +1000) /450) *10^4; plot([log10(VG) ,log10(J) ,VG=.1. .10]); J1:=subs(VG=VG+.01,J);
alpha_grain := ((log10(evalf (J1) *10^(-2))-log10(evalf (J) *10^(-2))) / (log10(VG+.01) - log10(VG)),VG=2.8..4.5);
plot(alpha_grain, title="Alpha of a ZnO grain versus Grain Boundary Voltage") ;VG:=(V/N);
JJLN:=sum(N*J* (sqrt(s*2*Pi) ) ^(-1) * (exp(-1/2* ( (log(N) - B) /sqrt(s) )^2)) ,N=round(B-
5*sqrt(s))..round(B+5*sqrt(s))); L:=20; P:=0.1; JJLN; JJ1LN: =subs (V=V+1 , JJLN);
ALN201 :=(log10(evalf (JJ1LN) *10^(-2))-log10(evalf(JJLN)*10^(-2)))/(log10(V+1)-log10(V));
P; L; plot(ALN201,V=0. .1000, title="Alpha LogNormal of the ZnO varistor versus the Applied Voltage");
B:=L1*(1+sum(P1^n, n=1. .infinity)); sn:=B*sum(P1^n,n=1. .infinity) ;readlib(log10);
J:=10^((tanh(50*log10(VG)-28))*4.5-
5.5) *10^( (500*log10(VG) +1000) /450) *10^4; J1:=subs(VG=VG+.01,J) ;VG:=(V/N); J;
JJN:=sum(J* (sqrt(sn*2*Pi) ) ^(-1) * (exp(-1/2* ((NB)/sqrt(sn))^2)) ,N=round(B5*sqrt(sn)) . .round(B+5*sqrt (sn)));L1:=20; P1:=0.1;JJN; JJN1:=subs(V=V+1,JJN); AN201 :=(log10(evalf (JJN1) *10^(-2))-log10(evalf(JJN)*10^(-2)))/(log10(V+1)-log10(V)); P1; L1; plot(AN201,V=0. .1000, title="Alpha of the Normal ZnO varistor versus the Applied Voltage"); P;L;plot([ALN201,AN201], V=40. .120, color=[red ,green ], style=[line]);
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