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Microstructure Characteristics of ZnO Varistors Simulated by Voronoi Network

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The Voronoi network can be used to effectively simulate the microstructure of ZnO varistors. The nonuniformity in microstructure of simulated ZnO varistor can be changed by setting different disorder degree of Voronoi network. In the region of disorder degree larger than 3 where the simulated microstructures are similar to those the actual ones of ZnO varistors, a chaotic phenomenon exists in the microstructure characteristics. This chaotic property can simulate the original behavior of nonuniformity of electrical characteristics caused by microstructures of ZnO varistors.

Key words : ZnO varistor, Voronoi network, Microstructural characteristics, Chaos phenomenon, Disorder degree

I. Introduction

ZnO varistors are polycrystalline ceramic devices which are widely used as surge arresters in power system and as surge suppressors in electronic system. They exhibit highly nonlinear current-voltage (*I-V*) characteristics, when the applied voltage exceeds the electrical breakdown voltage, empirically, $I=kV^{\alpha}$, the nonlinearity coefficient α can reach 60 or higher.

However, the microstructure of the ZnO varistor is highly nonuniform, the electrical properties of each grain boundary microjunctions are not identical.[1-5] A bulk varistor is a complex multijunction device composed of large numbers of both ohmic and nonlinear elements connected in a random network. As the overall properties of the varistor can be determined by the collective response of all the grain boundaries, electrical properties of ZnO varistors depend not only on the boundaries themselves but also on the properties of the grains and their topological arrangement in three dimensions.[6]

The influence of the geometry and topology of the granular microstructure, as well as the properties and the distribution of the different types of microjunctions on the features of bulk varistor devices, is an important and interesting problem. The behavior of varistors cannot be adequately described by simply averaging the characteristics of individual grain boundaries. On the other hand, the breakdown voltage of a bulk varistor, for example, is determined by the tail of the grain size distribution.[7-9]

Simulating calculation can effectively analyze the influences of different factors on the global electrical characteristics, influential factors include grain size, barrier voltage and nonlinearity coefficient of single grain boundary, and the effect of the distributions of these factors which are related to the nonuniformities of ZnO varistors, can be considered easily.

Calculating the effect of various microstructural variation on the *I-V* characteristics of varistors is difficult, due to the underlying complexity of the possible electrical transport paths through the microstructure when the grain boundaries have a nonlinearity, connected to each other in both parallel and series, depending in detail on the microstructure.

Several methods had been introduced to calculate the effect of nonuniformity of microstructure (grain size) and electrical characteristics (boundary barrier voltage and nonlinearity coefficient) on the global properties of ZnO varistors.[10,11,13] All these methods considered the ZnO varistors as simple electric circuits, and ZnO grain as a standard circle-, square- or rectangle-shape. We know ZnO varistor is a complicated network consisting of ZnO grains and grain boundaries in parallel and in series, and the connecting effect of ZnO grains and grain boundaries in horizontal direction is neglected

But the development of computer and calculation methods makes it possible to analyze the whole complicated network of ZnO varistors. More recently, Bartkowiak *et al.*[8,12] described the electrical properties of polycrystalline ZnO varistors by a two-dimensional Voronoi network model. The grains are represented by space-filling Voronoi polygons of different sizes and shapes with a variable number of neighbors. The geometry and topology of Voronoi networks closely resemble to those of found in grain growth from random nucleation sites. The stimulating results are particularly appropriate for the experimental results obtained with surface electrode patterns. Bartkowiak *et al.*[12] thought Voronoi networks can provide a realistic model of polycrystalline materials.[12] But the important phenomena of microstructure characteristics of ZnO varistors simulated by Voronoi network were not discussed.

We think that the Voronoi network can effectively simulate the actual microstructure of ZnO varistors, and consider the connecting effect of ZnO grains and grain boundaries in the whole network, which is a great progress in simulation of ZnO varistors. So, it is used to simulate the microstructure of ZnO varistors in this paper, and the important chaotic phenomenon of microstructure of ZnO varistors simulated by Voronoi network is discussed.

II. Voronoi Network Model of ZnO Varistors

A Voronoi polygon structure comprises randomly generated polygons that cover a spatial region with no gaps between adjacent polygons. This structure is generated using an algorithm that the spatial region to be partitioned around a random set of geometric centers in such a way that each polygon represents the region closer to a given center than any other center. As the structure consists of flat-sided polyhedra perfectly packed together, there is an interdependence in the shape between any polygon and its neighbours.[14,15] Voronoi network are formed by intersecting perpendicular bisectors of lines connecting neighboring seeds. The distribution of the geometric centers determines the granular geometry. When using different initial distributions of geometric centers, different granular geometries can be generated.

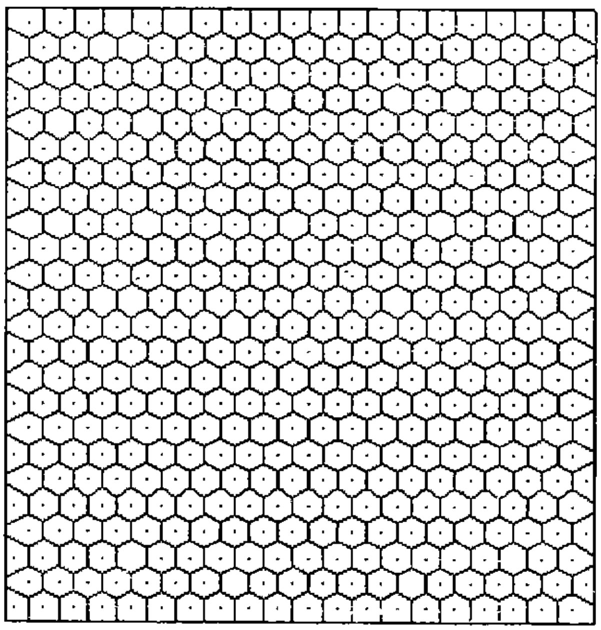
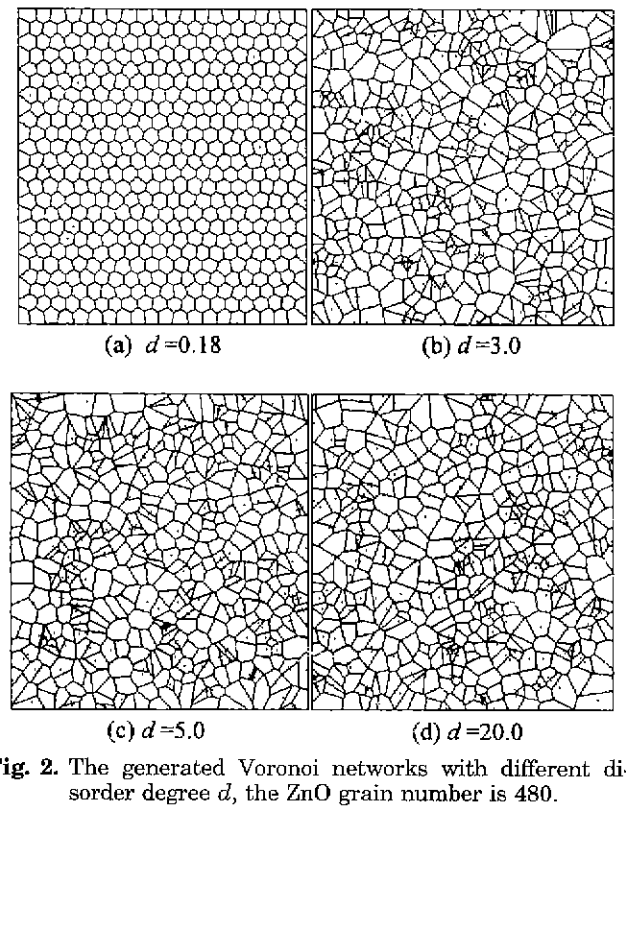


Fig. 1. The standard microstructure of ZnO varistor.

The nonuniformity of Voronoi network can be regulated by changing the disorder degree If the seed points are arranged in the same clearance $L=1$, disorder is introduced by displacing individual seeds within a disk of radius d around their original seed positions, where d is defined in unit of the distance between nearest neighbors in the triangular lattice and is called as disorder degree. Physically, Voronoi network can be interpreted as a two-dimensions growth process starting simultaneously at all nucleation seeds and proceeding in the plane at constant uniform rate until two approaching growth fronts reach each other. Therefore, the geometry and topology of Voronoi network closely resemble to those of found in grain growth from random nucleation sites.[5] So, it can effectively simulate the disordered structures of ZnO varistors and the nonuniformity of ZnO varistors can be simulated by changing the disorder degree. Priolo *et al.*[16] used the Voronoi network to simulate electromigration in random grain boundary network

When Voronoi network is used to simulate the microstructures of ZnO varistors, each cell represents a ZnO grain, and each edge of the length l_{ij} shared by neighboring cells i and j corresponds to a grain boundary. Bartkowiak and Mahan[8] first used the disordered two-dimensional Voronoi network to simulate the polycrystalline structure of ZnO varistors.

As ZnO grain has an wurtzite structure, the ideal microstructure of ZnO varistor consists of standard hexagons which corresponds to disorder degree d=0 (seen in



(a) d=0.18 (b) d=3.0

(c) d=5.0 (d) d=20.0

Fig. 2. The generated Voronoi networks with different disorder degree d, the ZnO grain number is 480.

Fig. 3. The Microstructure of ZnO varistor.

Fig. 1). When the disorder degree increases, the microstructure changes. ZnO varistors simulated by Voronoi networks with different disorder degrees are generated which is shown in Fig. 2. As discussed by Priolo *et al.*,[16] we observed the lattice becomes topologically disordered when the disorder degree above $d=(\sqrt{3}-1)/4=0.183$. When the disorder degree is larger than 3, the generated Voronoi network is very similar to the actual microstructure of ZnO varistor, and the topological structure can be regarded as generated from a fully random distribution of seed points, the SEM photo of actual ZnO varistor is shown in Fig. 3.

III. Chaos Phenomena of Microstructures of ZnO Varistors Simulated by Voronoi Network

The nonuniformity of microstructure of ZnO varistors is measured by the average grain sizes and standard deviations. Here we used the average equivalent-circle-diameter transformed from the area of an irregular shape and respective standard deviations as the average grain sizes $\overline{D}$ and standard deviations σ, because they can be automatically calculated by computer. The average grain sizes and standard deviations may affect the nonuniformities of electrical characteristics of ZnO varistors.

The nonuniformity of microstructure generated by Voronoi network at random disorder degree is easy to be obtained. A series of microstructures of ZnO varistors are generated at randomly selected disorder degrees by Voronoi networks, and the measured results are listed in Table 1. Here all the average grain sizes $\overline{D}$ of ZnO varistors simulated by Voronoi networks with different disorder degree are equal to 1. The relationships between the disorder degree d and the respective standard deviation σ, and the respective maximum grain size, are shown in Fig. 4 and Fig. 5, respectively. When disorder degree of Voronoi network is less than 3, the standard deviation and maximum grain size increase with disorder degree. But when disorder degree is higher than 3,

Table 1. The Respective Standard Deviation $\sigma/\overline{D}$, Respective Maximum Grain size $D_m/\overline{D}$ and Minimum Grain Boundary Number n_{gb} of all Paths Between two Electrodes Under Different Disorder Degree d

d	$\sigma/\overline{D}$	$D_m/\overline{D}$	n_{gb}
0	0.0209	1.0900	23
0.18	0.0374	1.1149	23
	0.0374	1.1153	23
0.30	0.0624	1.2337	22
	0 0626	1.2367	22
0 50	0.1024	1.3218	21
	0.1029	1.3283	21
1.0	0.1855	1.5444	21
	0.1889	1.5711	21
2.0	0 2436	1.5941	20
	0 2517	1.6450	20
3.0	0.2537	1.8985	19
	0.2538	1.8986	18
4.0	0.2739	2.0273	16
	0.2845	2.1066	18
5.0	0.2693	2.0778	16
	0.2795	2.1564	15
6.0	0.2543	1.6661	18
	0.2628	1.7219	14
	0.2735	1.8206	15
	0.2885	1.9242	16
	0.2863	2.0526	17
7.0	0.2735	2.1335	14
	0.2841	2.0916	17
8.0	0.2713	1.7665	16
	0.2643	1.7810	18
	0 2735	1.8216	14
	0.2818	1.8345	16
	0.2722	1.8506	16
	0.2801	2.0916	17
10 0	0.2703	1.7877	17
	0.2805	1.8559	16
15.0	0.2640	1.7242	15
	0.2736	1.7867	18
20.0	0.2733	1.8840	18
	0.2841	1 9576	14

there is an interesting phenomenon, under different disorder degree, the standard deviation changes irregularly in a range between 0.271 and 0.285, and maximum grain size changes between 1.666 and 2.156. Their average values are 0.271 and 1.911, respectively in the region where disorder degree is higher than 3. We observed that the standard deviations and maximum grain sizes of different microstructures arbitrarily generated with the same disorder degree change very small when the disorder degree is less than 3, but wide variations with the increase of disorder degree. They are very different and changes in their respective ranges shown above, when disorder degree is higher than 3 (Table 2). It is a kind of chaos phenomena according to the des-

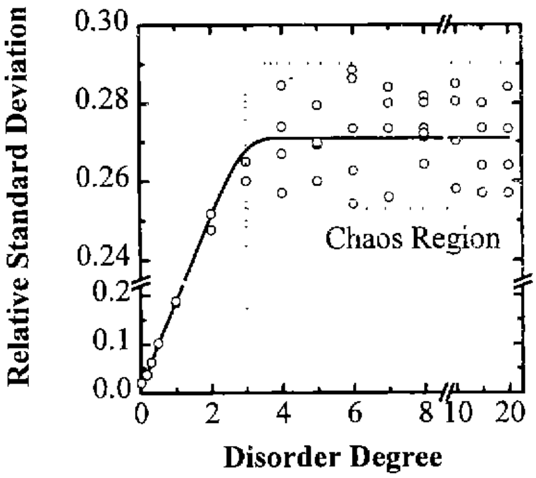


Fig. 4. The relationship between the disorder degree d and the respective relative standard deviation $\sigma/\bar{D}$ The average value of relative standard deviation is 0.271 in chaos region.

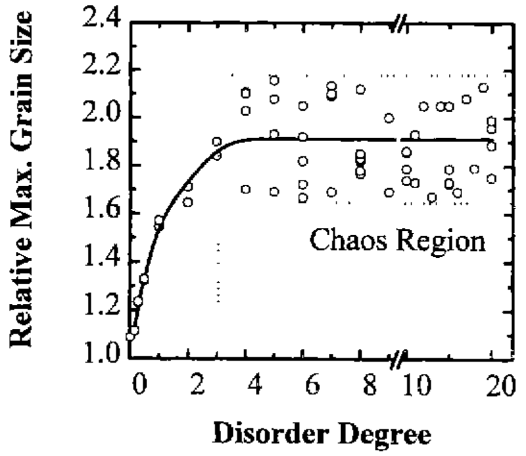


Fig. 5. The relationship between the disorder degree d and the respective relative maximum grain size $D_m/\bar{D}$.

Talbe 2. The Numbers of Different Grain Shapes in ZnO Varistors Generated by Voronoi Network with Different Disorder Degree

Disorder Degree d	0	0.18	0.30	1.0	2.0	5.0	8.0	20.0
triangle	0	0	0	6	9	6	4	3
qudrilateral	12	13	4	62	72	58	62	58
pentagon	50	49	65	138	127	154	148	152
hexagon	418	418	408	138	131	125	134	135
heptagon	0	0	3	82	85	90	85	78
octagon	0	0	0	40	33	34	28	36
nonagon	0	0	0	7	22	9	14	16
decagon	0	0	0	7	1	0	4	2
undecagon	0	0	0	0	0	3	1	0
dodecagon	0	0	0	0	0	0	0	0
tridecagon	0	0	0	0	0	1	0	0

cription to chaos by Gleick[17] and Lorenz,[16] and we define the region with disorder degree higher than 3 as chaos region.

As we think that the chaos phenomena of distributions of standard deviation and maximum grain size are caused by the grain size distribution, we analyzed the distributions of grain sizes of microstructures generated by Voronoi network under different disorder degrees. Grain sizes under different disorder degree have standard normal distribution. There is a trend that the peaked number becomes lower, and the distribution range becomes wider with the increase of disorder degree as seen in Fig. 6. When the disorder degree is larger than 3, the distributions under different disorder degree become irregular, but vary in a range shown in Fig. 6. A chaos phenomenon exists in the grain size distribution when disorder degree is larger than 3, which leads to the chaotic distributions of the standard deviation of grain size and the maximum grain size

The nonuniformity of electrical characteristics of ZnO varistors exists in the same varistor or different varistors. The influences of the nonuniformity of microstructure and grain boundary in the same varistor on the global electrical characteristics can be simulated by changing the disorder degree and setting different kind of barrier.[12] But the nonuniformity or deviation of electrical characteristics existing among different varistors still has not been simulated. We think it can be effectively considered by the chaotic property of ZnO microstructure simulated by Voronoi network with the disorder degree $d > 3.0$.

When a voltage is applied to the ZnO varistor, the current always looks for the path with minimum number of grain boundaries to pass through the ZnO varistor, the path with minimum number of grain boundaries will de-

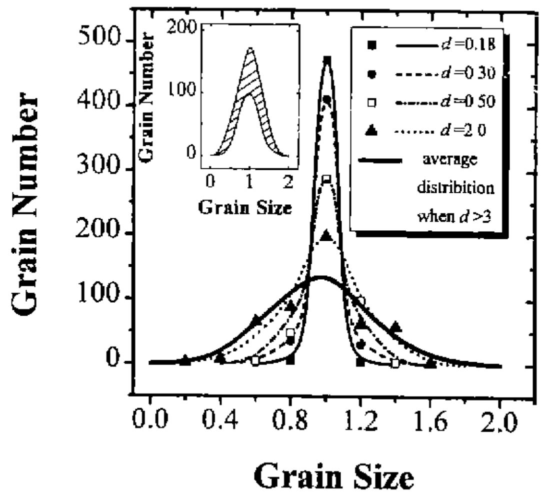


Fig. 6. The grain size distribution of ZnO varistors simulated by Voronoi network with different disorder degree. The small figure on the left upper corner is the distribution region of grain sizes when $d > 3$.

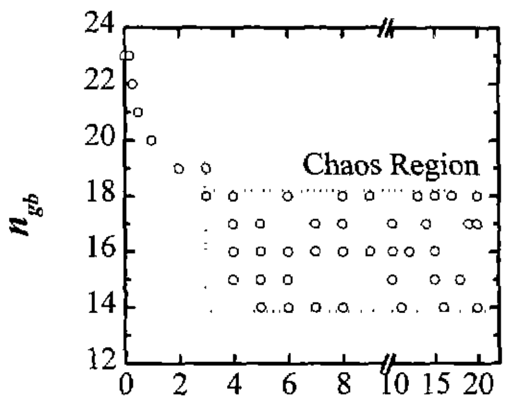


Fig. 7. The minimum grain boundary number n_{gb} between two electrodes of ZnO varistors simulated by Voronoi network.

termine the breakdown voltage and the global *I-V* characteristic of ZnO varistor. The minimum grain boundary number among all paths between two electrodes under different disorder degree is shown in Fig. 7. We observed that the minimum grain boundary number among all paths between two electrodes keeps the same when disorder degree is less than 0.18, then it increase with the increase of disorder degree in the disorder degree region $0.18 < d < 3.0$, but it becomes irregular when disorder degree $d > 3.0$. We analyzed the minimum grain boundary number in the region where $d > 3.0$ by changing the disorder degree and changing the microstructure in the same disorder degree, and observed that the minimum grain boundary number has a chaotic state, which changes irregularly in the range between 14 and 18. The chaotic state of the minimum grain boundary number among all paths between two electrodes leads to the nonuniformity of breakdown voltages among different ZnO varistors. The chaotic state of the minimum grain boundary number states that the nonuniformity of electrical characteristics existing among different varistors can be simulated by Voronoi network.

The numbers of different grain shapes in ZnO varistors generated by Voronoi network with different disorder degree are shown in Table 2. We observed the number of hexagon grain decreases and the numbers of other polygons increase with the increase of disorder degree When the disorder degree $d > 3$, the numbers of different grain shapes in ZnO varistors generated at the same disorder degree or different disorder degree are different, the numbers of different kind of polygons change irregularly in respective fixed ranges. There still are chaotic properties in the changes of different grain shapes

From the analysis above, we observed that the nonuniformity of microstructure of ZnO varistors can be effectively simulated by Voronoi network, which will lead to correct simulation to the nonuniformity of global electrical characteristics of ZnO varistors. And we can make statistical analysis on the global electrical characteristics of ZnO varistors based on the chaotic property of microstructures simulated by Voronoi network.

As discussed above, the microstructures of actual ZnO varistors are in the region with disorder degree $d > 3$. The chaos phenomenon states the nonuniformity of microstructures of ZnO varistors is an original property, which leads to the intrinsic property of electrical characteristical nonuniformity of ZnO varistors.

IV. Conclusions

Voronoi network is used to simulate the microstructures of ZnO varistors, grains are represented by space-filling Voronoi polygons of different sizes and shapes with a variable number of neighbors. The geometry and topology of Voronoi networks closely resemble those found in grain growth of ZnO varistors from random nucleation sites. The nonuniformity of microstructure of simulated ZnO varistor can be changed by setting different disorder degree of Voronoi network. In the region of disorder degree larger than 3, the simulated microstructures are similar to the actual ones of ZnO varistors, there is a chaotic phenomena existing in the microstructure characteristics, it states that the nonuniformity of microstructures of ZnO varistors is an original property. This chaotic property can simulate the intrinsic behavior of nonuniformity of global electrical characteristics caused by microstructures of ZnO varistors.

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