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The Influence of Dy_2O_3 doping on the Electrical Properties of ZnO-Based Varistor

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Abstract
ZnO is a ceramic material which tends to intrinsically form as an n-type semiconductor material. In this paper, the effect of Dy2O3 doping on the grain size and the electrical properties of ZnO-based varistor has been investigated, where we studied the I-V nonlinear coefficient behavior, the breakdown voltage, the potential gradient, leakage current, voltage per grain boundary before and after doping with Dy2O3 at concentration of 10-3 mol% and sintering temperature of 1050, 1100, and 1150oC.
Keywords: ZnO varistor, Dy2O3 doping, electrical properties.

1. Introduction
Zinc oxide (ZnO) varistors which mean (variable resistors) are polycrystalline ceramic devices exhibiting highly nonlinear (nonohmic) electrical behavior and greater energy absorption capabilities. The fabrication of ZnO varistors is done by mixing semiconducting ZnO powder with other oxides powders such as Bi, Co, Mn, Ni,Sb and Pr, and subjecting the powder mixture to conventional ceramic processing and sintering techniques, the sintering results in a polycrystalline ceramic with a singular grain boundary property which produces the nonlinear current-voltage (I-V) characteristics of the device[1,2]. Microstructurally, the ZnO varistors are comprised of semiconducting n-type ZnO grains, surrounded by very thin insulating inter granular layers[1,3].

In operation, a varistor is connected between the power source and ground, when the electric field exceeds the switching field, the surge is carried away through the varistor, thus protecting the circuit or the power utility[4].

The nonlinear V–I characteristics of ZnO varistors ceramics are attributed to a double Schottky barrier (DSB) formed to the grain boundaries, which are essentially formed by a segregation of varistors forming oxides [3,5,6].

The nonlinear current-voltage (IV) characteristics described by the following relations.

$$J = KE^{\alpha}$$

$$\alpha = \frac{\log(J_2/J_1)}{\log(E_2/E)}$$

Or

$$I = KV^{\alpha}$$

$$\alpha = \frac{\log(I_2/I_1)}{\log(V_2/V_1)}$$

$$\alpha = \frac{1}{log(V_{10mA}/V_{1mA})}$$

Where J is the electrical current density, K is a constant that depends on the microstructure , $J_2 = 1\ mA/cm^2$, $J_1 = 0.1\ mA/cm^2$, E is the potential gradient α is the nonlinear coefficient, and $I_2 = 1\ mA$, $I_1 = 0.1\ mA$, $V_{1\ mA}$ and $V_{0.1mA}$ represent the voltage at 1 and 0.1mA respectively[4,6,7,8,9,10].

Bi-based zinc oxide varistors have been studied in different aspects although Bi-based zinc oxide varistors show good nonlinear properties, Bi_2O_3 easily reacts with some metals used in preparing multilayer chip nonlinear varistors[11].

2. Sample Preparation
Varistor samples were fabricated by the conventional ceramic fabrication procedure. Appropriate amounts of raw chemicals were used in proportions of [(95-X) mol% ZnO, 0.5 mol % Bi_2O_3, 2.5 mol % Sb_2O_3, 0.5 mol % Co_3O_4, 0.5 mol % Cr_2O_3, 0.5 mol % NiO and 0.5 mol % MnO_2, where (X=0.001, 0.005, 0.01 mol % Dy_2O_3)]. Raw materials were mixed by high-energy using magnetic stirrer with magnetic bar in glass container (dry mixing) for 24 hours.

The mixture was calcined at 600 ^oC in air for 2 hours with heating rate equals to *5^{o}C/min* ,then the mixture crashed and remiled, after 3 wt % polyvinyl alcohol (PVA) binder addition, the powder was uniaxially pressed into discs of 15 mm in diameter at a pressure of 25 MPa. The discs were sintered in air at 1050, 1100, and 1150°C with heating rate of *5°C/min* for 2 hours.

The breakdown voltage measured at the current of 1mA, the potential gradient $E_{1mA} = V_{1mA}/D$, where D the sample thickness, and the leakage current (I_L) was determined at 0.75 V_{1mA}.

The voltage per grain boundary, $V_{gb} = V_b(\frac{d}{D})$, where V_b the breakdown voltage, d the average grain size and D is the thickness of the sample.

These properties measured and accomplished by using An instrument manually designed in the laboratory and DC Power supplies of ranged from 0~1500 volt ,50 mA model (HIRANUMA EP-1500, Japan) and (PHYWE High voltage 0... 10 kV, 2 mA) , where the current changed manually during the tests.

3. Results and Discussion

Figure 1 shows the phase composition of the samples. It is observed that ZnO is the main phase and $Zn_7Sb_2O_{12}$ is the secondary phase.

It must be observed that the X-ray diffraction peaks of the rare earth oxide are very weak, because of the small concentration rate of this oxide in the samples. But the effect of them on the electrical properties is obvious.

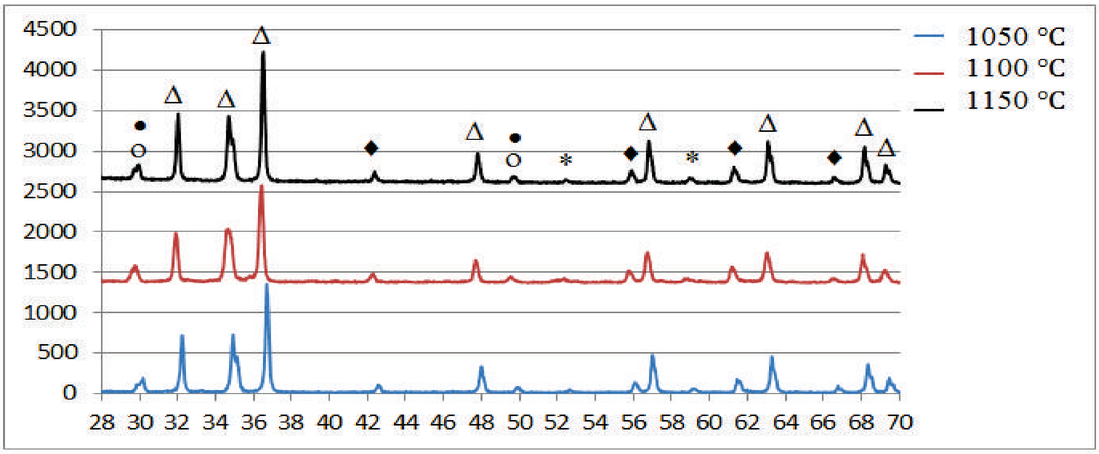

Figure 1. XRD data of the samples, where o Bi_2O_3, • Sb_2O_3, ♦ $Zn_7Sb_2O_{12}$, Δ ZnO, and ∗ Dy_2O_3.

Table.1 and Figure.2. Illustrate effect of Dy_2O_3 doping on the grain size, simply we can observe that the doping with Dy_2O_3 increase the grain size of the varistor.

Table 1. Grain size values of samples before and after Dy_2O_3 doping.

Sample	Grain size before doping *nm*	Sample	Grain size after doping *nm*	Sintering Temperature °C
S1	43.04	S4	44.15	1050
S2	48.40	S5	50.21	1100
S3	1225	S6	1305	1150

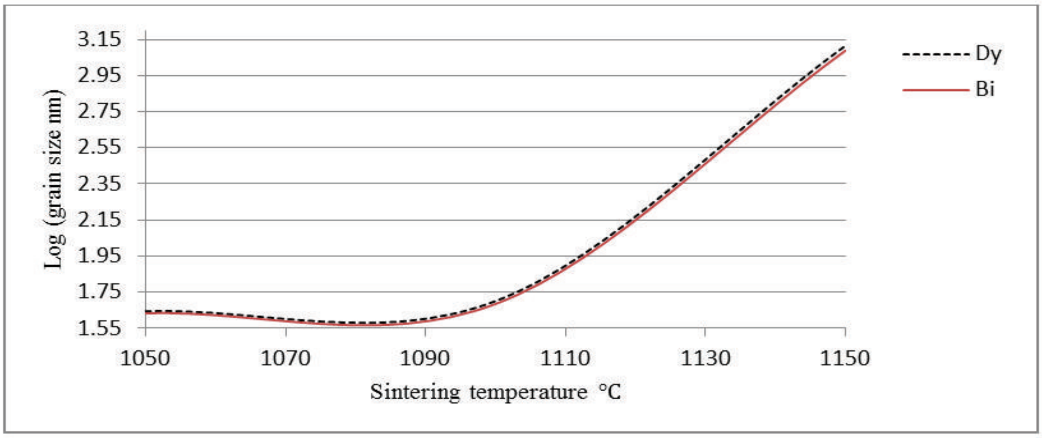

Figure 2. Grain size Log values curves before and after Dy_2O_3 doping

Table. 2, 3 and Figure. 3, illustrate the electrical properties behavior versus the sintering temperature, before and after doping with Dy_2O_3.

As we can see the doping with Dy_2O_3 increase the non-linear coefficient, breakdown voltage, and potential gradient , which directly proportional with sintering temperature until it reach the maximum value at 1100°C, then it begins to decrease, while voltage per grain boundary continues to increase.

Decreasing of non-linear coefficient starts soon after sintering temperature of 1100°C.

The behavior of electrical characteristic with temperature have one meaning that the volatilization of spinal phase ($Zn_7Sb_2O_{11}$) and Bi_2O_3 phase which represent the insulating layer between the conducting grains of ZnO and in addition to increasing the grain size, therefore that leads to decreasing the number of the junctions (Schottky barriers) between the grains.

Table 2. Electrical characteristic of samples before Dy_2O_3 doping.

Sample	S.T (°C)	α	B.V $volt$	P.G V/mm	L.C μA	V_{gb} volt
S1	1050	4.46	2320	1532	33	0.092
S2	1100	6.46	950.4	668.9	56.4	0.155
S3	1150	5.5	724.8	505.6	69.1	0.296

Table 3. Electrical characteristic of samples after Dy_2O_3 doping, where S.T sintering temperature, B.V, breakdown voltage, P.G potential gradient, L.C leakage current, and V_{gb} voltage per grain boundary.

Sample	S.T (°C)	α	B.V $volt$	P.G V/mm	L.C μA	V_{gb} volt
S4	1050	16.74	3340	1977	44.4	0.936
S5	1100	30.08	3920	2565	50.5	1.52
S6	1150	25.13	3632	2508	46.3	2.27

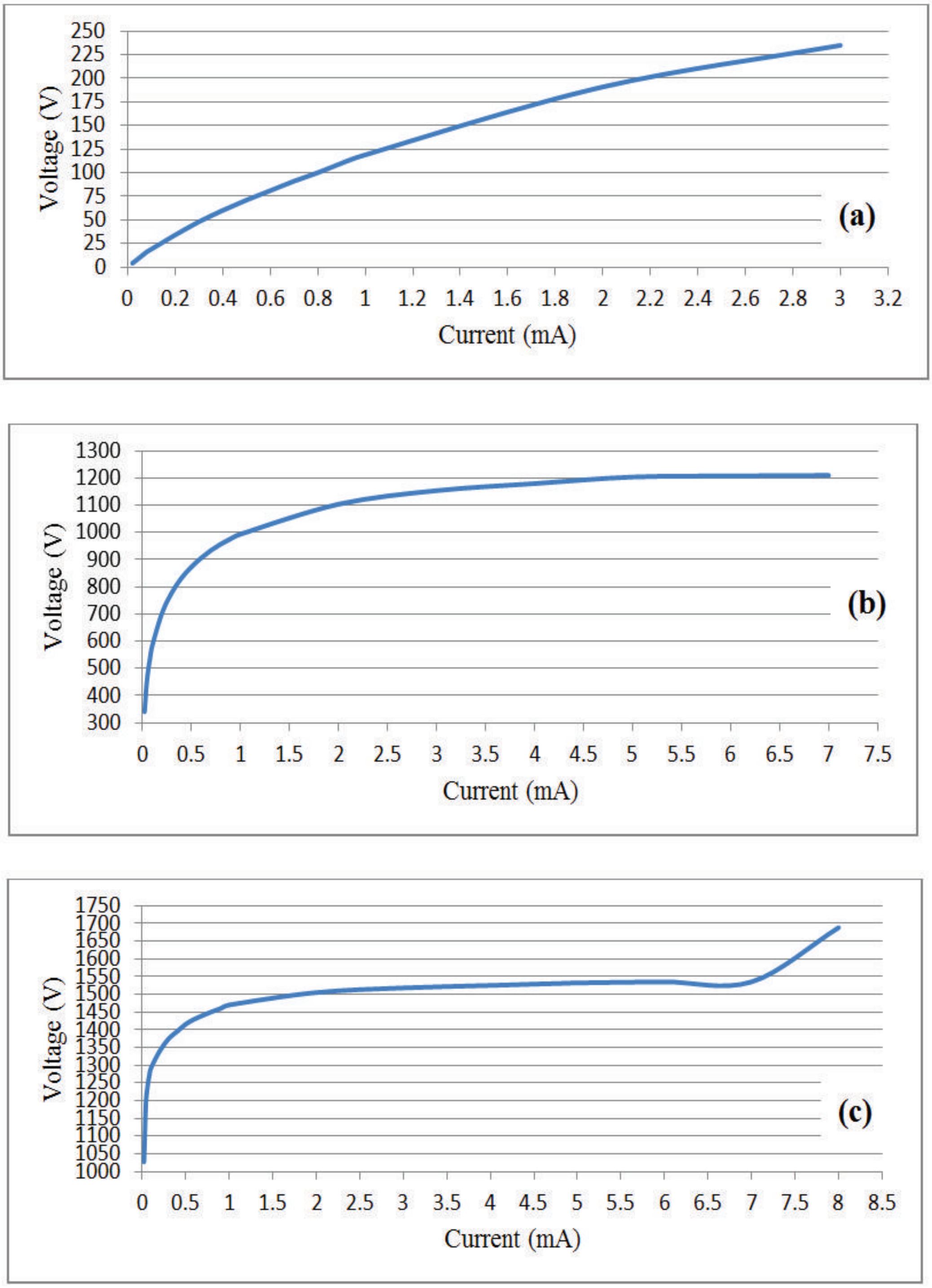

Figure 3. I-V characteristic curve of, (a) ZnO pure, (b) before Dy_2O_3 doping, (c) after Dy_2O_3 doping.

4. Conclusion

ZnO has ohmic (linear) behavior, and good basic properties enable it to be a base several devices, and one of the most important usages is a Surge protection device (varistor). ZnO pure is useless or unprofitable as a varistor, while the doping of ZnO with some metal oxides changes the behavior of ZnO varistor from linear to non-linear. The doping of ZnO varistor with rare earth oxides elevates the magnitude of improvement, and efficiency of it.

The increasing of rare earth oxides concentration improves the electrical properties of the varistor especially the non-linear coefficient which represents the fundamental feature in varistor work.

The raising of sintering temperature enhanced the electrical properties until 1100^oC, but more than 1100^oC decreases the efficiency of varistor, because of Bi_2O_3 and spinal ($Zn_7Sb_2O_{11}$) phases volatilization which decrease the insulation barrier between the ZnO grains.

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