International Journal on
"Technical and Physical Problems of Engineering"
(IJTPE)
Published by International Organization of IOTPE

ISSN 2077-3528
IJTPE Journal
www.iotpe.com
ijtpe@iotpe.com

March 2024	Issue 58	Volume 16	Number 1	Pages 138-143

ANALYSIS OF THE MAIN PARAMETERS OF ZnO POLYMER COMPOSITES WITH DIFFERENT THICKNESSES

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Abstract- The article is devoted to the analysis of the main parameters of ZnO-polymer composites with different thicknesses. In the article, study the variation of dielectric permeability, which are the main parameters of ZnO-polymer composite varistors with different thicknesses, nonlinearity of volt-ampere characteristic (β) and opening voltage (U_{cr}) under normal conditions and under the influence of external factors. For this purpose, samples with thicknesses of 40, 180 and 300 µm and different volume percentages (30÷60%) were prepared and their corresponding characteristics were measured. It should be noted that various experiments were conducted by changing the type of additives and polymers that make up the ZnO phase of the composite. As a result of the experiments, it was found that the preparation of composites with different thicknesses, as well as the types of additives and polymers significantly affect their electrophysical parameters.The opening voltage (U_{cr}) and the coefficient of non-linearity (β) of the volt-ampere characteristic, which are the main parameters of the composites, were determined. The most effective nonlinearity was observed in composites with a thickness of 180 µm. The main parameters of the composites after modification and before modification were also measured in the work. In all cases, the dielectric constant (ε) increased as the volume percentage of the composites increased.

Keywords: ZnO, Varistors, Composites, Impurities, Potential Barriers, Nonlinearities, Critical Voltages, Thickness, Dielectric Constant, Modification.

1. INTRODUCTION

Zinc oxide is widely used in modern technology. Being an optically transparent wide-gap semiconductor, zinc oxide is used for the production of components for high-power semiconductor devices (thyristors, varistors), UV filters, and solar cells. Relative chemical and biological inertness make it possible to use ZnO as components of drugs.

In high-displacement technology, ZnO is widely used to limit the displacement and flow backs that arise as a result of various reasons. The key element that determines the properties of a ZnO varistor is the turn-on voltage and non-linearity of its volt-ampere characteristic. The non-linearity of the volt-ampere characteristic of elements based on ZnO changes in the interval of 50-70. ZnO varistors are connected in series-parallel and placed in the dielectric ground. One of the important factors when using varistors is their parameter stability. For this purpose, electrothermal aging of varistors is carried out: varistors are heated to 403-423 K under the influence of an electric field, and then cooled to room temperature. This technological operation is repeated until the non-linearity of the opening voltage of the varistor and the stabilization of the resistance up to opening voltage [1-5].

It should be noted that when selecting semiconductor ceramic materials for the development of varistors with different functions, the conductivity of their crystallites, the width of the barrier zone, the shortness of electric currents, and the parameters of the potential difference that can occur at the boundary between amorphous and crystalline phases should be taken into account.

One of the main factors of importance in the development of composite varistors based on Si-polymer and ZnO-polymer is the determination of the dependence of the varistor parameters of the semiconductor ceramic phases of the composites Si and ZnO on the width of their band gap. The width of the forbidden zone depends on the parameters of the potential barrier formed at the crystal-amorphous phase boundary in SiЪ and ZnO materials [6-15]. It is shown in the literature that the width of the forbidden zone of SiC is 2.2 eV. Therefore, untreated SiC is not a conductive material.This effect ensures that the resistance after the turn-on voltage of the SiC varistor is high, and the non-linearity of the varistor's volt-ampere characteristic is reduced. Therefore, studying the effect of SiC deposition on both SiC itself and the characteristics of composite varistors made on the basis of Si-polymer is one of the fundamental issues in this field [16-23].

Registered: 230522 Article Info: 1711 Accepted: 240224
Reserved: 240117 Available: 240331

The influence of additives on the electrophysical properties of primary SiC has been sufficiently studied [24, 25]. It is assumed that p-type semiconductor materials are formed when Si is doped with elements of the fifth group, and the conductivity of varistors based on such materials after the opening voltage depends on the doped concentration. A p-type semiconductor is obtained when Si is dissolved with second and third group elements. As a varistor, p-type Si is used more often. It is known from the available experimental results [4] that in the process of joining p-type Si, electrons are easily transferred from one crystallite to another and a polycrystalline structure is formed. The number of contacts in SiC depends on the size of the crystallites. Due to the current passing through the contacts, the contacts stabilize continuously and new current lines are connected. Between the crystallites there is a very thin transition. According to the Pool-Frenkel effect, the tunneling of electric currents can occur among transition processes. ZnO is a semiconductor compound and belongs to type $A^2 B^6$ [26-33].

The width of its blocking zone is greater than the corresponding parameter of Si. Therefore, as the resistance after the opening strain of the initial ZnO that has not been refined is greater, its non-linearity coefficient should be small. However, despite the large width of the band gap, the stoichiometry of ZnO is defined due to the lack of oxygen in the cooking process, and therefore it has n-type semiconductor. Between the crystallites of ZnO there is a very thin insulating region that ensures the formation of the varistor effect. A ZnO varistor with a cross-sectional area of 1 cm^2 withstands current pulses with an amplitude of several kA and can reduce high voltage spikes to the operating voltage.

Regarding the development of composite varistors, it is important to choose which type of semiconductor (p or n-type) and to study the influence of the bandgap zone width of the semiconductor phase on the characteristics of the varistor.

The main purpose of the work is to study the variation of dielectric permeability, which are the main parameters of ZnO-polymer composite varistors with different thicknesses, nonlinearity of volt-ampere characteristic (β) and opening voltage (U_{cr}) under normal conditions and under the influence of external factors. For this purpose, samples with thicknesses of 40, 180 and 300 μm and different volume percentages (30÷60%) were prepared and their corresponding characteristics were measured.

2. METHOD OF ANALYSIS

The synthesis of used ZnO and SiC varistors is carried out by ceramic method. The synthesis process is due to diffusion between solid particles, resulting in the formation of a solid phase. Higher temperatures are used to increase the diffusion of particles. Thus, ceramic synthesis is sometimes carried out at a temperature of 2300 K [16]. In order to facilitate mutual diffusion of particles, they are first mechanically crushed as much as possible. During crushing methods used in industry, particles with a size of 10-60 μm are usually obtained.

The crushed mass is pressed under high pressure to improve the contact of these particles. However, since ceramic synthesis is diffusion-based and low penetration rate and time consuming for one atom the particles to completely diffuse into the other. So, if the size of the particle is 10 μm and the elementary lattice parameter of the primary substance is 10 Å = 10^{-7} cm = 10^{-3} μm, then the atom needs to pass 10000 elementary lattices during diffusion of that particle from one side to the other. It sometimes takes more than 10 hours for an atom to travel such a path. In industry, ceramic synthesis is usually carried out in a relatively short time (2÷10 hours).

Therefore, ceramic synthesis often proceeds only around the contact boundaries of the particles. The diffusion process begins at the contact boundary of the particles. At this time, an intermediate phase is formed at the boundary. If the speed of the initial diffusion process depends on the chemical nature of the particles in contact, the temperature of synthesis, after the formation of the intermediate layer, it also depends on the thickness of that layer and the resistance of atoms and ions to diffusion dynamics. Often, the reaction rate decreases exponentially depending on the thickness of the intermediate phase.

Thus, the chemical nature of the intermediate phase, state of aggregate, crystal structure, penetration properties accept a high effect every kinetics about ceramic synthesis. If diffusion permeability of intermediate phases to ions involved in the diffusion is large, then the reaction will go faster. Usually, when preparing the charge, they use a polymorphous form of the substance so that it undergoes a polymorphous transition during the synthesis. The intermediate phase formed at the temperature of the process can also be in a liquid state. If the intermediate phase is liquid, the reaction is fast. In practice, in order to speed up the ceramic synthesis, oxide or halide compounds, which melt more easily than the initial batch, are added. One of the factors influencing the kinetics of ceramic synthesis is the presence of polymorphous forms of substances involved in the synthesis. If one of the components undergoes a polymorph transition in the synthesis temperature interval, the speed of the synthesis process will increase significantly at that transition temperature. This phenomenon, called Tendal effect, is widely used in ceramic synthesis [7-17].

In order for the parameters of varistors to remain stable, it is an important condition to protect them from moisture. The final operation of making varistors is to attach electrodes to their surface. This operation is carried out as follows: after the chemical cleaning of the surface of the elements, the electrodes are applied and baked from the paste on the surface. To protect the electrodes from the environment, their surface is varnished. To increase the non-linearity of the volt-ampere characteristic of varistors, they use pulsed and pulsed firing methods [24-25]. Figure 1 shows the volt-ampere characteristics of the ZnO+Pe composite with different thicknesses.

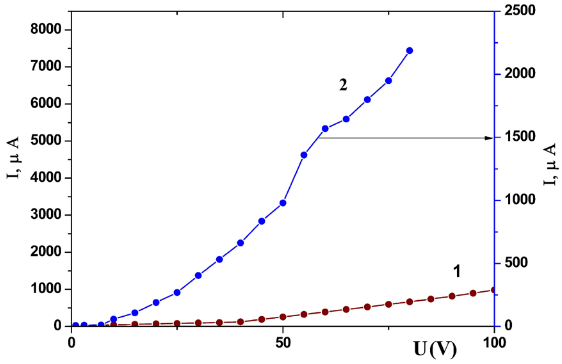

Figure 1. Volt-ampere characteristics of 50%ZnO+50%Pe composite with different thickness 1- h = 40 μm, 2- h = 300 μm, h is thickness of the sample [7]

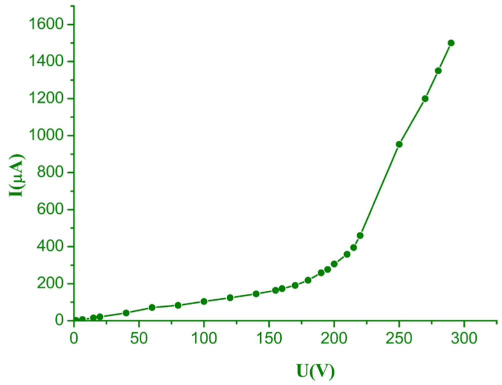

Figure 2. Volt-ampere characteristics of 50%ZnO+50%Pe composite, h =180 μm [7]

Figure 2 shows the volt-ampere characteristic of the ZnO+Pe composite with a thickness of 180 μm. It can be seen from Figures 1 and 2 that the thickness of the samples has a great effect on the volt-ampere characteristic. At a certain thickness of the composite varistor, the opening voltage (U_{cr}) significantly decreases and the nonlinear coefficient (β) increases.

According to the experimental results, the main properties of the synthesized multilayer varistors depend on the development of electron-ion exchange processes in the polymer phase, and the height of the potential barrier at the phase boundary is determined by the volume fraction and size of the main structural element of ZnO ceramics [30].

Tables 1 and 2 also show the main parameters of the volt-ampere characteristic, the turn-on voltage (U_{cr}) and the non-linearity coefficient (β) of the ZnO-polymer based composite varistors with various additives.

Table 1. Critical voltage (U_{cr}) and nonlinearity factor (β) of composite varistors based on ZnO-PVDF with different additives [29]

Composites	Critical tensions (U_{cr}) V and nonlinearity coefficient (β)			
	ZnO (with additives ZrO_2) +F2M		ZnO (with additives Al_2O_3) +F2M	
	U_{cr} V	β	U_{cr} V	β
30%C+70% PVDF	215	6.8	U=140	4.86
35%C+65% PVDF	198	6.6	U=137	5
40%C+60% PVDF	182	6.35	U=125	7
50%C+50% PVDF	150	6.2	U=120	7.6
60%C+40% PVDF	130	5.7	U=117	8.7

Table 2. Critical voltage (U_{cr}) of composite varistors based on ZnO-PE with different additives [29]

Composite samples	Critical tensions (U_{cr}), V	
	ZnO (with additives ZrO_2) +Pe	ZnO (with additives Al_2O_3) +PE
30%C+70%PE	U=600	U=140
40%C+60%PE	U=310	U=150
50%C+50%PE	U=250	U=190
60%C+40%PE	U=200	U=120

The volt-ampere characteristics of these composites were measured and the critical voltage (U_{cr}) and non-linearity (β) of the volt-ampere characteristic were determined.

In the first approximation, we believe that changes in U_{op} and β depending on the structure and properties (polarity) are associated with interfacial processes at the polymer-ZnO interface. The experimental results show received are the size particles of the inorganic phase is influencing the CVC of the composite varistor. By at certain (constant) thickness of the composite varistor, the operation voltage decreases markedly, and the nonlinearity coefficient increases. Our various findings from experiments show the importance of impact of electric discharge plasma on the polymer-zinc oxide composite results in an important modification that permittivity and the concentration of local levels at the interface of the composite [16].

In figure 3 has shown the dielectric permittivity with the volume content of zinc oxide for samples treated electric discharge plasma's influence. It can be seen that for all volumetric contents, the treatment with electricity discharge plasma results in a notice able a rise in. This parameter increases with increasing duration of the discharge. In figure shows the adjustments to the coefficient of nonlinearity (β) of the volt-ampere characteristic composite before and then their working in an electric gas discharge for various volumetric contents of the ZnO phase. It can be seen that plasma treatment significantly increases the values of β.

As we mentioned, one of the main properties of composite varistors is its critical voltage (U_{cr}) and the relationship between this voltage and the filler's volume proportion. The fact that the critical voltage depends on the polymer matrix's characteristics (polarity, non-polarity) shows that this effect is not related to the processes taking place in the ceramic phase. It can be considered that this effect is related to the electronic processes at the boundary between the ceramic phase and the polymer phase. Indeed, at the contact boundary between polymer and ceramic, electron exchange takes place due to the difference in the output work of polymer and ceramic.

As a result of electron exchange, a boundary potential similar to the Schottky barrier is created at the boundary. It can be considered that since the output work of the polymer is greater than the output work of the ceramic, electrons pass from the ceramic to the polymer. A positive charge is created in the ceramic corresponding to each passing electron.

Therefore, a layer consisting of charges in the ceramic and transferred electrons in the polymer, i.e., at the boundary, a potential barrier develops. As the heights and widths of the potential barrier decreases, the tunneling effect-based electron transfer at the polymer-ceramic interface will be smaller [16-23].

Figure 3 shows the dependence of the dielectric constant on the volume percentage of the filler.

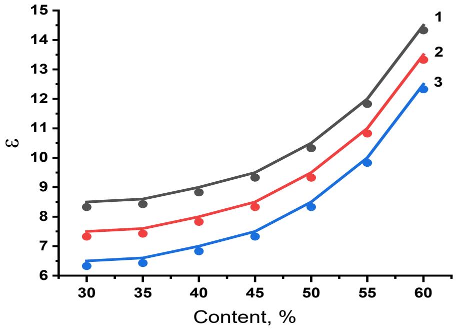

Figure 3. Dependence dielectric constant of the volume content filler in the ZnO-high-pressure polyethylene composite and treatment by electric discharge (1- untreated, 2- treated for 3 min, 3- for 10 min) [16]

It can be seen from Figure 3 that before and after the discharge of electric gas, the dielectric strength increases linearly depending on the volume percentage and nonlinearly after about 40% volume percentage. Let's say that the value of the dielectric strength will increase in all percentages as the effect of the gas release time increases.

It should be noted that the change in the polarization process in the composite as a result of the impact of the gas release process leads to an increase in the dielectric constant of the composite.

Let's note that the opening voltage of acidic ZnO-polymer composite varistors is inversely proportional to the dielectric constant of the interphase boundary. The dielectric strength of the composite depends on the dielectric strength of its components and the percentage of the composites according to the Maxwell-Wagner formula. According to experimental studies, the dielectric strength and Volt-Farad characteristics of the transition layer formed as a result of the interaction of ZnO particles (filler phase) with the polymer are different from the corresponding characteristics of the polymer matrix and alkaline ZnO. If such an effect is possible, it can be said that the dielectric strength of the composite varistor as a whole change according to a very complex law.

The experimental results showed that as the volume percentage of ceramics increases in thin-film composite varistors based on various semiconductors (ZnO, Si, GaAs, InAs) and polar and non-polar polymers (Pe, PP, PVDF), the ceramic particles are closer to each other. are located at a distance, and as a result, the thickness of the polymer layer between them decreases [6, 7]. This, in turn, leads to the creation of local levels in these layers, that is, the polarization of this layer. This process causes a change in the electrophysical parameters of the composites. As a result, the specific volume resistance of the composites decreases, and the conductivity and dielectric permeability increase [26-33].

3. CONCLUSIONS

The analysis obtained results indicates the dependence at operational characteristics of multilayer varistor on the development of the electron-ion exchange processes at polymer phase. The size of the possible obstacle at phase boundary primarily depends on the volume percentage and size of the main structural element of ZnO ceramics. Value and resistance of the potential barrier between them ceramic particle and polymers decreases due to the increase in dielectric permeability. According to the conducted experiments, as the volume percentage of the filler increases, the value of the dielectric permeability of the composite increases.

It was found that the value of the dielectric constant in percent of the whole volume will increase as the impact period of the gas release process increases. It should be noted that the electrophysical properties of the composite are highly dependent on the duration of the impact of the gas release process. Electrophysical parameters also change as the duration of exposure increases. This is related to the fact that ozone, nitrogen oxides, etc., generated with increased energy during the gas release process. The concentration of such substances increases and their impact on the composite increases. In addition, during the gas release process, the degree of crystallization of the polymer changes - that is, it can increase or decrease.

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