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A Study of The Main Electrophysical Parameters of Semiconductor - Polymer Based Composite Varistors

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ABSTRACT

The main electrophysical parameters of composite varistors made on the basis of filled zinc oxide (ZnO), monocrystalline silicon (Si), gallium arsenide (GaAs), indium arsenide (InAs) and various polymers were studied in this work. In the article, the sample preparation process is described, interphase interaction is discussed. The nonlinearity coefficient (β) and opening voltages (U_{op}) of volt-ampere characteristics in filled ZnO, monocrystalline Si ceramic semiconductors and polymer-based composite varistors were determined. The volt-ampere characteristics of monocrystalline Si, GaAs and InAs and polymer-based composites were also measured. The shape of the potential hole in the mentioned composites has been determined. It was found that the opening voltage and the nonlinearity of the volt-ampere characteristic of polymer-semiconductor composites mainly depend on the properties of the 3rd phase. According to the experiment, it was found that as the filler volume percentage increases in ZnO, monocrystalline Si, GaAs and InAs and polymer-based composites, the βincreases in all samples, and the U_{op} decreases. Depending on the type of dispersant, the opening voltages of the composites are different. Thus, in ZnO-polymer-based composites with additives, this voltage varies between 130-220 V, and in monocrystalline Si, GaAs and InAs and polymer-based composites, it varies between 5-50 V. The analysis of the shape of the potential hole in composites based on monocrystalline Si, GaAs, and InAs has shown that the value of the forbidden zone in the composites decreases, and the value of the potential barrier decreases sharply.

Yarı İletken-Polimer Esaslı Kompozit Varistörlerin Ana Elektrofiziksel Parametrelerinin İncelenmesi

MAKALE BİLGİSİ

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Anahtar Kelimeler:
ZnO
Monokristal Si
Kompozit varistörler
Seramik
Polimer
Açılış voltajı

ÖZET

Bu çalışmada katkılı çinko oksit (ZnO), monokristalin silikon (Si), galyum arsenit (GaAs), indiyum arsenit (InAs) ve çeşitli polimerler temelinde yapılan kompozit varistörlerin ana elektrofiziksel parametreleri incelenmiştir. Makalede numune hazırlama süreci anlatılmakta, fazlar arası etkileşim tartışılmaktadır. Katkılı ZnO, monokristal Si seramik yarı iletkenler ve polimer bazlı kompozit varistörlerdeki volt-amper özelliklerinin doğrusal olmama katsayısı (β) ve açılış gerilimleri (U_{op}) belirlendi. Monokristal Si, GaAs ve InAs ile polimer bazlı kompozitlerin volt-amper özellikleri de ölçüldü. Söz konusu kompozitlerdeki potansiyel deliğin şekli belirlenmiştir. Polimer-yarı iletken kompozitlerin açılış geriliminin ve volt-amper karakteristiğinin doğrusal olmama durumunun esas olarak 3.fazın özelliklerine bağlı olduğu bulunmuştur. Deneye göre ZnO, monokristal Si, GaAs ve InAs ile polimer bazlı kompozitlerde dolgu hacim yüzdesi arttıkça tüm numunelerde β'ninarttığı, U_{op}'ninazaldığı tespit edildi. Dağıtıcının türüne bağlı olarak kompozitlerin açılış gerilimleri farklıdır. Dolayısıyla ZnO-polimer bazlı katkı maddeli kompozitlerde bu voltaj 130-220 V arasında değişirken, monokristal Si, GaAs ve InAs ile polimer bazlı kompozitlerde 5-50 V arasında değişmektedir. Monokristal Si, GaAs ve InAs bazlı kompozitlerdeki potansiyel delik, kompozitlerdeki yasak bölge değerinin azaldığını, potansiyel bariyer değerinin ise keskin bir şekilde azaldığını göstermiştir.

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1. INTRODUCTION (GİRİŞ)

It is known that composite materials are a material made of two or more constituent materials with significantly different physical and chemical properties and having different properties when combined. Composite materials are composed of several components-usually a plastic base (matrix) and additional strength, stiffness, lightness, etc. it consists of an amplifier-a filler. It is possible to develop composites with different properties depending on the direction, field of use, composition.

It should be noted that defense of microelectronics, electronic devices and their functional components from switching and lightning voltages is one of the important problems. Various types of varistors are used in the electronics industry all over the world to guard electrical networks and electronic devices from extreme electrical impulses of the desired type. In this regard, ceramic varistors based on ZnO and monocrystalline Si, which differ from other materials by a number of advantages, have found a wider field of application. It should be considered that recently, one of the promising directions for the improvement of protecting devices and components is the manufacture of two- and multi-phase composites based upon ceramic varistors. By preparing these composites on the basis of ceramic phase and polymers, it is potential to obtain composite varistors that are cheaper and of higher quality than those available so far, both in high-voltage equipment and in low-voltage devices of power engineering [1-7].

By injecting certain additives into ZnO ceramics, it is possible to synthesize it with polymer materials and make thin-film composite varistors and make a wide variety of them expediently. Currently, the issues of renewing the composition of composite materials, developing cheap, economical and effective composite materials and wide application of these composites in the industry are very relevant. For this purpose, various additives were added to the ZnO phase [8].

Polymer-based thin-film composite varistors, apart from the simplicity of the manufacturing technology, also have advantages in terms of some operating parameters. By changing the temperature-time dependence of the crystallization of the polymer matrix, it is possible to adjust the properties and stability of the parameters of the composite varistors. By adding additives to the ceramic phase, it is possible to significantly change the surface activity of polymer-based composites, as well as the nonlinearity coefficient of the volt-ampere characteristic and the opening voltage. It should be noted that low temperature, very small weight and geometric dimensions, non-linearity coefficient and appropriate variation of the opening voltage of polymer-based thin-film composite varistors are of great practical importance.

The occurrence of effects observed in composites is mainly related to the formation of a potential barrier at the interphase boundary. In turn, the formation of various properties in composites depends greatly on the electrophysical parameters of the potential barrier formed at the interphase boundary and the processes occurring at the polymer-filler boundary. Therefore, by changing the composition of composites and the properties of phases, it is possible to create different effects in them and create more effective devices for different fields of technology based on composites.

It should be noted that the practical importance of the work is that the composites created on the basis of wide-band polymers and ceramic materials can be used as overvoltage limiters, electronic switches, memory elements, etc. can be used as Also, the obtained polymer-based thin-film composite varistors can be applied in various fields of electrical engineering by appropriately changing the opening voltage.

The purpose of the study is to determine the basic electrophysical parameters, i.e., opening voltage (U_{op}) and nonlinearity coefficient (β) of the volt-ampere characteristic in composite varistors made on the basis of filled ZnO, monocrystalline Si, GaAs, InAs and various polymers. For this purpose, the synthesis process of composites, the opening voltage (U_{op}), the nonlinearity factor (β) and the shape of the potential hole of composite varistors made on the basis of additive ZnO, monocrystalline Si, GaAs, InAs and various polymers were determined.

2. EXPERIMENTAL PART (DENEYSEL BÖLÜM)

Synthesis of composite varistors based on filled ZnO, monocrystalline Si, GaAs, InAs and various polymers is carried out by ceramic method [9-10]. Higher temperatures are used to increase particle diffusion. Thus, ceramic synthesis is sometimes carried out at a temperature of 2300 K.
1000 grams of ceramics are weighed and crushed in a porcelain ball mill (trademark FRITSCH) to a size of 60 μm or less. Then, samples with a height of 10 mm and a diameter of 20 mm are obtained from this mixture by pressing at a pressure of 40 tons. After that, for synthesis, the samples are placed in the furnace and heated to 900 ºC at a rate of 150 °C/s, and to a temperature of 1250 °C at a rate of 200 °C/s. The process of synthesis of pressed samples is carried out for 2 hours at a temperature of 1250 °C, in an air environment. After turning off the oven, the samples are cooled for 7-8 hours.

To facilitate mutual diffusion of the particles, they are first mechanically crushed as small as possible. During the comminution 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. Nevertheless, ceramic synthesis is diffusive, and when the diffusion rate is small, it takes a long time for the atom of one particle to completely diffuse into the other. So, if the size of the particle is 10μm and the elementary lattice parameter of the primary substance is $10Å=10^{-7}cm =10^{-3}$ μm, then the atom (ion) needs to pass 10000 elementary lattices during the diffusion of that particle from one side to the other. It sometimes takes 10 hours or more for an atom to travel such a path.

Ceramic synthesis in industry is usually carried out in a relatively short time (2-10 hours) [5]. Therefore, ceramic synthesis often occurs only around the contact boundaries of the particles, and the diffusion process begins at the contact boundary of the particles. At this time, an intermediate phase is formed at the border (Figure 1). If the speed of the initial diffusion process depends on the chemical nature of the particles in contact, the synthesis temperature, after the intermediate layer is formed, it also depends on the thickness of that layer and the resistance of atoms and ions to the diffusion dynamics. Often, the reaction rate decreases exponentially depending on the thickness of the intermediate phase.

The components of the composite varistors used in the work are based on filled ZnO, monocrystalline Si, GaAs, InAs semiconductor ceramics and non-polar polyethylene (PE), polypropylene (PP) and polar polyvinylidene fluoride (PVDF) polymers.

It should be noted that the Co_2O_3, MnO_2, Cr_2O_3, Sb_2O_3 oxides present as additives in the synthesized ZnO varistor react with ZnO and form spinel-type $ZnCr_2O_4$ (a= 8.32Å), $Zn_7Sb_2O_{12}$ (a= 8.58Å), $Co_7Sb_2O_{12}$, (a = 8.55Å), $MnCr_2$ (a = 8.436Å), $CoCr_2O_4$ (a= 8.32Å), etc. forms compounds.

First of all, semiconductor ceramics and polymers are mixed together to obtain composites [11-13]. The mixing process is carried out in micromills, is a mixer device that ensures homogeneous mixing of polymer and reinforcing element. It should be noted that polymers with strong dielectric properties were used as the polymer phase [14,15]. Figure 1 shows the sample mixing process.

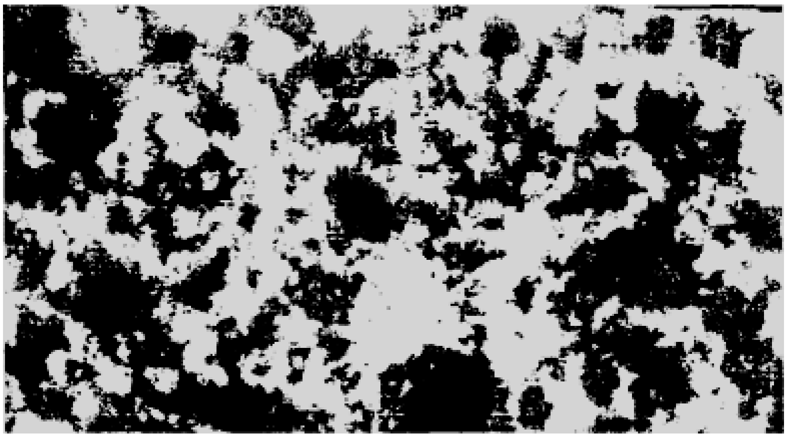


Figure 1.Sample mixing process (Numune karıştırma işlemi)

The individual components of the mixture were first dried in the form of powder, polymers at 413 K, and ZnO, Si, GaAs and InAs ceramics at 523 K. The structure and electrophysical parameters of individual components of the mixture depend on the demand placed on the varistor to be made on its basis [16].

It should be noted that it is of particular importance to obtain a homogeneous mixture. Because interphase interactions are important in the formation of the varistor effect in the semiconductor-polymer composite. Therefore, the equal distribution of the processes taking place at the interphase boundary in the volume of the sample significantly affects its electrophysical parameters [17-19].

For any material to be a varistor, it must have areas in its physical structure that have markedly different electrophysical, thermal, and physical properties (i.e., crystalline and amorphous phases).When composites with this basic requirement are considered, it is seen that polymer-ceramic and polymer-semiconductor composites meet this requirement [20]. Namely, the role of the crystalline phase is assumed by the ceramic or semiconductor, and the role of the amorphous phase is undertaken by the polymer matrix.However, many microscopic and structural studies expression that it is the third phase that powerfully impacts the development of the varistor effect in composite materials and the electrophysical properties, supramolecular structure and electronic events at the interphase boundary. This phase occurs at the polymer-ceramic and polymer-semiconductor boundaries.

Figure 2 shows the interaction between the third phase formed at the polymer-ceramic boundary in the ceramic-polymer composite.

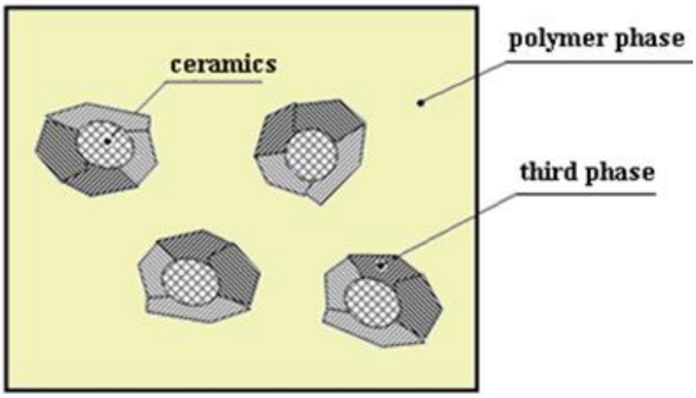


Figure 2. Interphase interactions (Fazlhararası etkileşimler)

It can be seen from Figure 2 that the 3rd phase is formed by the effect of the surface of inorganic particles. The nonlinearity of the opening voltage and volt-ampere characteristics of polymer-ceramic and polymer-semiconductor composites mainly depends on the properties of the 3rd phase.

It has recently been established that between the numerousidentified organic compounds and composites based on them, there is not a large class of multiphase materials with a very strange combination of properties: bipolar non-omic conductivity, heat resistance, sufficiently high mechanical and electrical strengths, and a symmetrical probable barrier on the phase boundary. Such materialsnamed active composites include, particularly, varistors based on polymers distributed by ZnO, monocrystalline Si, GaAs, InAs semiconductor ceramic particles [19]. A novel group of composite elements has been created, whose distinguishing feature is non-bipolar conductivity. Contrastingordinary non-linear devices, they do not cover asymmetric potential barriers and current-voltage characteristic (CVC). These materials have a wide scope, ranging from nanoelectronics, integrated circuits, to low-power switches for numerous goals [19].The prospect of polymer composites with bipolar conductivity is due to both wide functional capabilities (for example: in the function of energy-intensive and low-power varistors), and a relatively simple and low-temperature manufacturing technology. The obvious nonlinearity of conductivity and the symmetric nonlinear CVC property of polymer-semiconductor composites attract attention as an active composite for varistors with wide possibilities for varying the non-linearity coefficient of the current-voltage characteristic, operating voltage (U_{op}), and the temperature range of operation. By the gathering of experimental records, the model idea of the creation mechanism of the varistor impact and the conductivity of composites transformed from a model in which the main role was ascribed to the polymer sheetsamong ZnO and Si particles to a model wherein various grain-boundary properties in the ZnO, Si, GaAs and InAs particle itself and events on the boundary sheets of the polymer phase with involving particles of ZnO, Si, GaAs and InAs.

3. DISCUSSION OF RESULTS (SONUÇLARIN TARTIŞILMASI)

By means of the gathering of experimental records, the model idea of the formation mechanism of the varistor impact and the conductivity of composites transformed from a model in which the chief role was ascribed to the polymer sheetsamong ZnO, monocrystalline Si, GaAs and InAs particles to a model wherein various grain-boundary effects in the semiconductor ceramics particle itself and events on the boundary sheets of the polymer phase with involving particles of semiconductor ceramics.

Since the main parameters determining the varistor effect of varistor materials are the non-linearity of their volt-ampere characteristic (β) and opening voltage (U_{op}), analogous parameters of composite varistors have been determined.The graphs based on the obtained results are shown in Figure 3 and Figure 4.

Figure 3 shows the dependence of the non-linearity (β) of the volt-ampere characteristic in the filled ZnO-Pe composite varistor on the filler volume percentage.

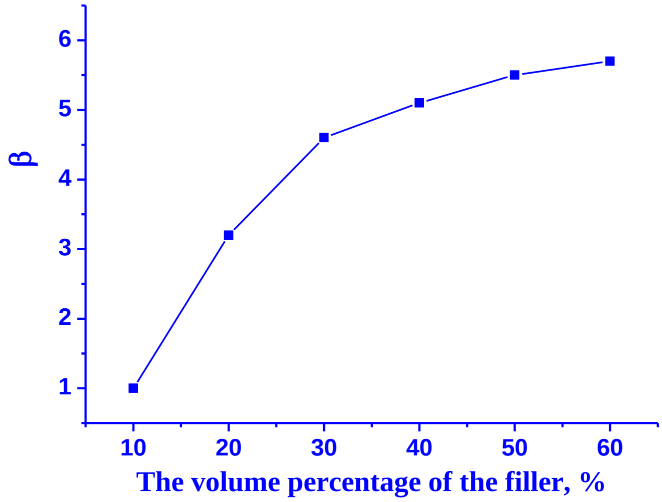


Figure 3. Dependence of the nonlinearity of the volt-ampere characteristic ZnO-Pe composite varistor on the filler volume percentage (Volt-amper karakteristiği ZnO-Pe kompozit varistörün doğrusal olmama durumunun dolgu hacmi yüzdesine bağımlılığı)

It is clear from Figure 3 that the non-linearity coefficient (β) of the composite varistor's volt-ampere characteristic increases significantly (β=1-5.5) as the volume fraction of the filler in the composite increases.

Figure 4 shows the dependency of the nonlinearity coefficient of monocrystalline Si-polymer composite varistors on the volume percentage of the disperser.

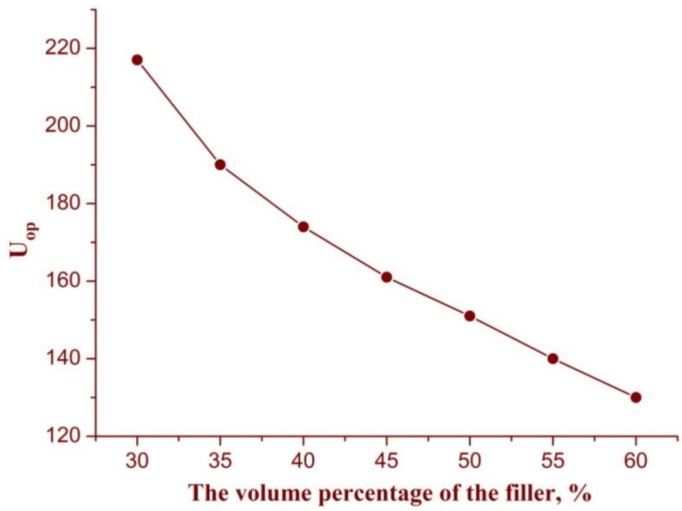


Figure 4. Dependence of the opening voltage in the ZnO-Pe composite varistor on the filler volume percentage (ZnO-Pe kompozit varistördeki açılış geriliminin dolgu hacmi yüzdesine bağımlılığı)

It can be seen from Figure 4 that in filled ZnO-Pe based composite varistors, as the filler volume percentage increases, the opening voltage value also increases.

Figure 6 shows the dependence of the opening voltage on the volume fraction of the filler in a monocrystalline Si-polymer varistor.

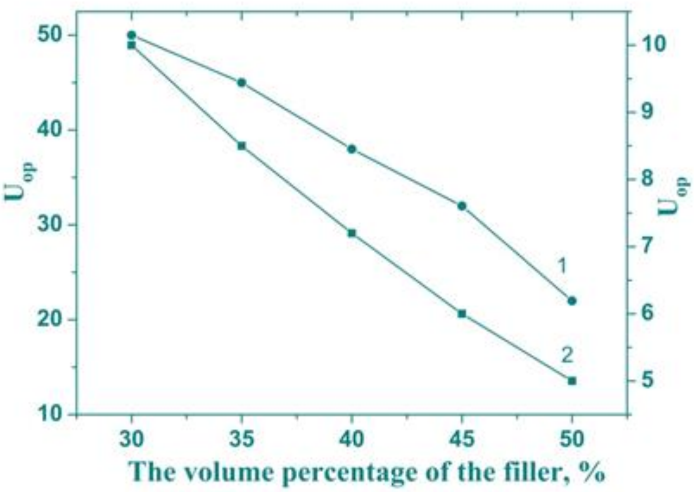


Figure 5. Dependence of the opening voltage on the volume percentage of the filler in monocrystalline Si-polymer based composites; 1-Si-PP; 2-Si – PVDF. (Monokristal Si-polimer bazlı kompozitlerde açılma voltajının dolgu maddesinin hacim yüzdesine bağlılığı)

It can be seen from Figure 6 that the opening voltage (U_{op}) of the composite varistor decreases as the volume fraction of the filler in the composite increases. The explanation of Figure 3-6 is as follows.

The interaction between phases in polymer composites depends the intermolecular structure of the polymer phase, the polarity of its macromolecule, and the surface movement of the dispersant particles. In turn, the interphase mutual influence ultimately affects the chief properties of the semiconductor-polymer varistor[20-30].

As we said, the key reason for the formation of the varistor influence in composites is the formation of a potential barrier at the boundary between the particles of semiconducting ZnO ceramics and the polymer phase and the possibility of its modulation (regulation) through the applied electric field. It can be considered that the parameters of the potential barrier, which determine the opening voltage of the polymer-ZnO varistor and the nonlinearity of the volt-ampere characteristic, depend on the electrophysical characteristics and chemical structure of the contacting surface of the phases. Additives injected into ZnO in certain amounts significantly affect the parameters of the ZnO-Pe composite varistor. Initially, we believe that the additives affect the chemical and physical structures of ZnO and ultimately change the electron-ion process at the boundary of the polymer and ZnO phases. Therefore, the opening voltage value of ZnO-polymer composite is higher than that of Si-polymer based composites.

Figure 6 shows the volt-ampere characteristics of composite varistors based on disperser and non-polar PP with different volume percentages.

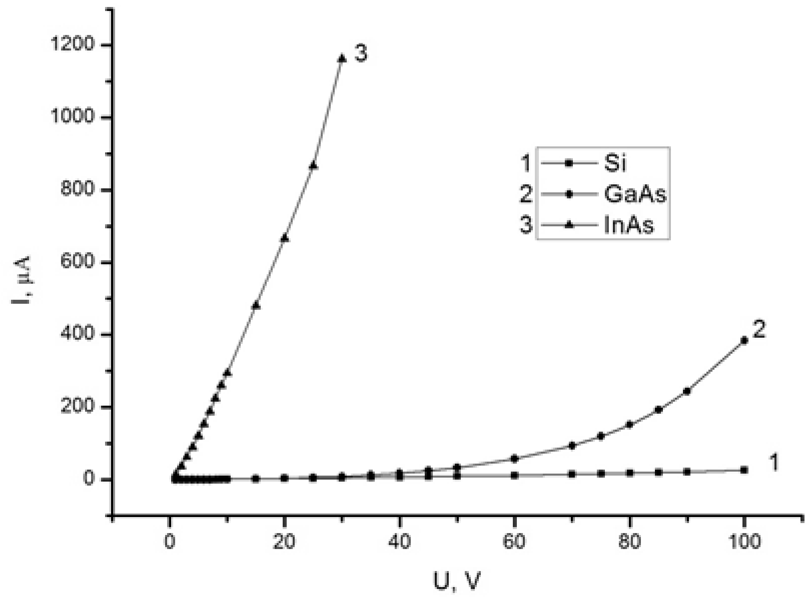


Figure 6. Volt-ampere characteristics of 50% semiconductor + 50% polypropylene based composites (%50 yarı iletken + %50 polipropilen bazlı kompozitlerin volt-amper özellikleri)

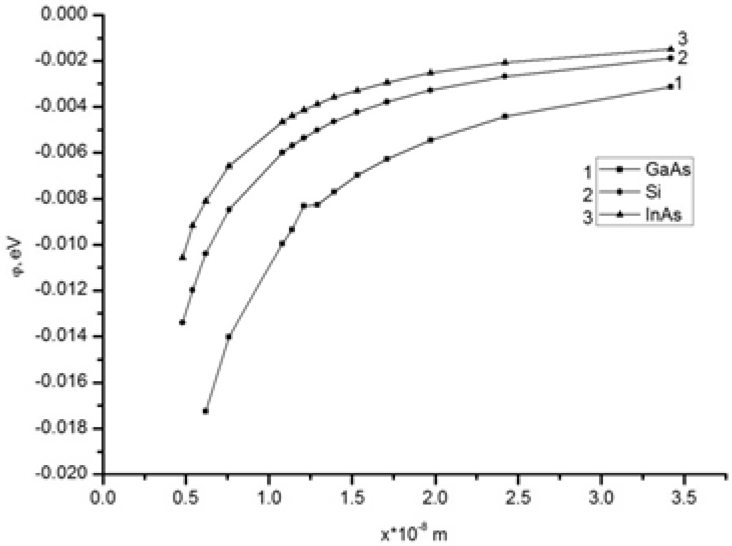
It can be seen from figure 6 that, depending on the type of dispersers, the opening voltages of varistors vary in the range of 50-5 V. From the analysis of the pictures, it can be seen that the influence of the components of the composites on the volt-ampere characteristics of the studied samples is evident.

The obtained results are explained from the point of view of the mechanism of heat transfer in polycrystalline structures. So, these mechanisms; a) tunneling of the potential barriers created due to the dielectric (polymer) nature between the dispersant and polymer particles; b) is the emission of electric currents from the gap between the polymer and semiconductor particles and the transfer of electric currents along the chain formed as a result of the direct contact of the dispersant particles in the composite. Another reason for observing the non-linearity in volt-ampere characterictic is the direct contact of neighboring particles formed during their cooking. Such a contact causes microheating due to electron emission from the sharp ends of the particles when the current passes through the sample.

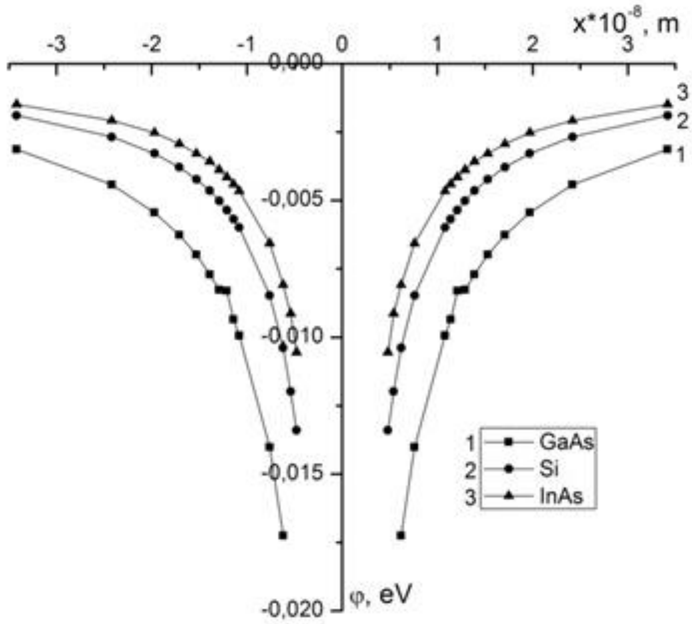
According to the work of [11-25], they say that the conductivity of the composite is a function of the average of the contacts corresponding to one particle. Also, based on the theory of electrical contacts, the flow of electric current between 2 conductors is possible not only by their direct contact, but also by the existence of a thin dielectric layer between them. In this case, the conductivity is created by tunneling through the potential barrier of the superconductors. In turn, the tunnel resistance is exponentially dependent on the value of the potential barrier [15].

In small percentages of the dispersant, the value of the probable barrier is high due to the high density of the polymer nature between the semiconductor particles. On the other hand, due to the exponential dependency of the tunnel resistance on the value of the probable barrier, the probability of the superconductors tunneling through the fire barrier decreases sharply, and the tunnel permeability will be small in small percentages of the disperser. Accordingly, the strength of the composite will be determined mainly by the strength of the polymer. In this part, the tolerance is of a sliding nature. With the increase of the volume percentage of the disperser, the thickness of the polymer layer between the semiconductor particles decreases and the mean value of the contacts between the particles rises, as a result, the strength of the potential difference and the value of the tunnel resistance decreases.

Figure 7a,b shows the shape of the potential hole in the positive(+) and negative(-) regions in different dispersant-based composites.



a)



b)

Figure 7. a - Shape of the potential hole in different dispersant-based composites,
b- Shape of the potential hole in the positive(+) and negative(-) region
(a - Farklı dağıtıcı bazlı kompozitlerdeki potansiyel deliğin şekli,
b- Pozitif(+) ve negatif(-) bölgedeki potansiyel deliğin şekli)

Figure 7a,b clearly shows the influence of the type of dispersant on the formation of the potential well in the composites. So, on the one hand, in all three types of semiconductor-based composites, the shape of the potential cross-section is similar, as the value of x increases, the value of φalso increases. On the other hand, the value of the forbidden zone decreases in the order of GaAs, Si and InAs, and the value of the strength of the potential barrier decreases sharply.

It should be noted that electron exchange takes place at the contact boundary between polymer and ceramic due to the different work functions of polymer and ceramic. As a result of electron exchange, a boundary potential similar to the Schottky barrier is generated at the boundary. It can be calculated that because the work function of the polymer is greater than the work function of the ceramic, electrons transfer from the ceramic to the polymer. Each night is positively charged in an electron-compatible ceramic. Therefore, a potential barrier is formed at the boundary, consisting of charges in ceramics, and electrons in polymer. Depending on the strength of the potential barrier, the boundary resistance changes. The smaller the height and width of the potential cross-section, the more electrons will be transferred at the polymer-ceramic interface based on the tunnel effect at smaller gaps [21-32].

4. CONCLUSIONS (SONUÇLAR)

Interphase interactions are important in the formation of varistor influence in semiconductor-polymer composite. In order to study the interphase interactions, one of the important issues is the determination of the main parameters that determine the varistor effect of composite varistors - the nonlinearity of their volt-ampere characteristic (β) and opening voltage (U_{op}), as well as the optimal sample percentages. For this purpose, analogous parameters of composite varistors are defined in the article. The homogeneity of composites obtained on the basis of additive ZnO, monocrystalline Si, GaAs and InAs ceramics and polymers was ensured. During the analysis of the studies, it was found that the opening voltage of the semiconductor-polymer composites and the nonlinearity of the volt-ampere characteristic mainly depend on the properties of the 3rd phase.

The obtained experimental results show that the non-linearity coefficient (β) of the volt-ampere characteristic of the composite varistors increases sufficiently as the volume fraction of the filler increases in both ZnO-polymer and monocrystalline Si-polymer based composites under the conditions of stability of all other electrophysical and geometric size factors, and the opening voltage decreases (U_{op}). It should be noted that the dispersion of polymer dielectrics with ceramic particles leads to the formation of a potential barrier at the interphase boundary, which leads to the formation of the varistor effect in the composite and its change under the influence of the electric field. Stabilization of electrons with available beams reduces the resistance of the varistor and the thickness of the potential barrier between the crystallites, which leads to a decrease in the opening voltage of the composites. A sufficiently large non-linearity ratio indicates the effectiveness of composite varistors.

It should be noted that the type of disperser is established based on the formation of a potential hole in composites. Thus, the shape of the potential barrier in different types of dispersant-based composites has a similar character. In the order of GaAs, Si and InAs, the value of the forbidden zone decreases, and the value of the potential barrier decreases sharply.

All the mentioned parameters ultimately affect the breakdown voltage (U_{op}) and nonlinearity coefficient (β) of the volt-ampere characteristic. The change of the opening principle in a wide interval and the variation of the nonlinearity coefficient allow the preparation of effective and high-quality composite varistors according to the purpose.

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