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TEMPERATURE DISTRIBUTION IN THE VICINITY OF DEFECTS IN SEMICONDUCTOR BARIUM-TITANATE-BASED CERAMICS

V. N. Shut and A. V. Gavrilov

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The temperature distribution in the region of defects (pores, voids, and large grains) in barium-titanate-based ceramics in the process of its heating by electric current has been investigated by numerical methods.

Introduction. In electrical engineering and radio electronics, cells with a positive temperature coefficient of resistance (PTCR) are widely used. Semiconductor ceramics based on barium titanate doped with rare-earth elements form the most important group of PTCR cells. This is due to the high value of the temperature coefficient of resistance and the possibility of varying the switching temperature and the resistance value of samples over a wide range [1]. However, $BaTiO_3$-based thermistors may fail by the delamination mechanism under the action of large current loads [2]. Large currents cause nonuniform heating of the posistors, as a result of which temperature stresses arise and lead to the appearance of cracks and fracture of ceramics (delamination into two equal halves in the plane parallel to the electrodes).

The temperature field is influenced by both external factors (heat exchange with the environment, applied voltage) and the properties of the ceramics proper (heat conduction, heat capacity, grain sizes, inclusions). It should be taken into account that barium-titanate-based cells in the process of switching to the high-resistance state are heated to above the Curie temperature (T_C). The ferroelectric phase transition of the first kind is accompanied not only by the appearance of the PTCR effect, but also by a stepwise change in the T_C range in many physical parameters of the material (dielectric constant, thermal expansion coefficient, heat capacity, Young modulus, heat conduction, etc.).

A number of authors [2, 3] investigated the temperature distribution in the depth of samples without taking into account the microstructure of the ceramics. The local temperature fields within the limits of one grain were also investigated [4, 5] since the grain has a high-resistance boundary layer and a low-resistance central region.

The general view of the temperature field should be influenced by defects (inclusions of the air phase in the form of pores and voids, inclusions of large grains) that inevitably take place in the production of ceramics. However, investigations of such a kind have not yet received proper attention. The aim of the present work is to investigate the temperature fields in the region of defects of semiconductor barium-titanate-based ceramics in the process of its heating by electric current.

Theoretical Model. The current technologies permit obtaining posistor ceramics with an average grain size of 5–10 μm. As was shown in [6], the temperature differential in a grain of size 10 μm does not exceed a few tenths of a degree (at a voltage of 220 V and a specific resistance over 0.4 Ω·m). Therefore, the consideration of posistor ceramics as a homogeneous material having individual inclusions of defects in the form of pores and voids, as well as of large grains, is quite justified. The validity of such an approach is confirmed by the investigation of the microstructure of posistors, whose typical photograph is given in Fig. 1. It is seen that against the background of fairly uniformly size-distributed grains there are voids of size over 20 μm and large grains.

Subsequently, ceramics with a grain size of 5–10 μm were treated as homogeneous. Grains and voids of sizes larger than the average grains size of ceramics (over 10 μm) were considered as defects. The temperature distribution in the posistor is described on the basis of the simultaneous solution of the equations of heat conduction

$$c\rho^* \frac{\partial T}{\partial t} - \nabla (\lambda \nabla T) = Q \tag{1}$$

Engineering Acoustics Institute, National Academy of Sciences of Belarus, 13 Lyudnikov Ave., Vitebsk, 210023, Belarus; email: shut@vitebsk.by. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 80, No. 4, pp. 181–185, July–August, 2007. Original article submitted January 21, 2006; revision submitted September 12, 2007.

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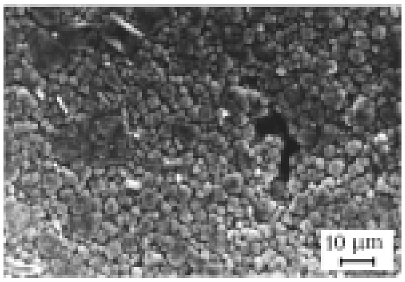


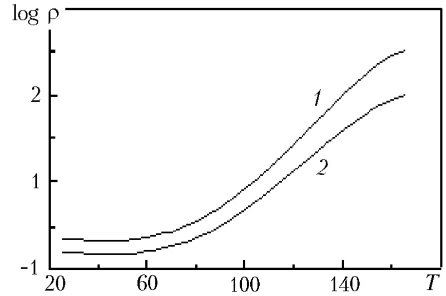


Fig. 1. Photograph of the microstructure of posistor ceramics.

Fig. 2. Temperature dependence of the specific resistance of protective posistor ceramics: 1) zero strength of the electric field; 2) electric field strength of 100 W/mm.

and electrical conduction

$$-\nabla\left(\frac{1}{\rho}\nabla\varphi\right)=0\,. \tag{2}$$

The boundary conditions for the heat conduction equation were chosen proceeding from the assumption that the thermal flow on the surface is proportional to the temperature difference between the surface and the environment (Newton's law)

$$\mathbf{n}\cdot(\lambda\nabla T)=\sigma\,(T_0-T)\,. \tag{3}$$

The boundary conditions for the electrical conduction equation were chosen proceeding from the assumption that the side surface of the posistor is insulated

$$\mathbf{n}\cdot\mathbf{I}=0\,, \tag{4}$$

and a potential difference of 220 V is applied to the electrodes

$$\varphi\Big|_{x=0}=0\,, \tag{5}$$

$$\varphi\Big|_{x=L}=220\,. \tag{6}$$

The above equations were solved by the finite element method.

The specific resistance of posistor ceramics P depends on the composition of the material, the technological conditions of its production, the sample temperature, and the voltage applied (varistor effect). The dependences of the specific resistance on the temperature and the electric field strength were obtained experimentally (Fig. 2) ("Monolitradiokeram" protective thermistors were investigated). The data obtained were approximated by mathematical expressions and used in solving Eqs. (1) and (2).

Results and Discussion. The location of a defect is schematically represented in Fig. 3. Figure 4 shows the results of modeling the temperature distribution in the region of a void of size 50 μm situated at a distance h = 50 μm from the side surface in the YZ (a) and XY (b) planes 2.5 msec after voltage was applied. As is seen from Fig. 4a, a higher temperature is observed around the void in the plane perpendicular to the current direction (in the YZ plane). And the local maximum thereby is near the side surface of the posistor (in the region of the local maximum of the current density and the E-vector). At the same time, the heat generation near the front and back surface of the defect (in the XY plane) is minimal (Fig. 4b).

Table 1 systematizes the data on the change in the maximum temperature differential $\Delta T_{\max}$ (difference between the local maximum and minimum) in the vicinity of voids of different sizes. It is seen that the maximum tem-

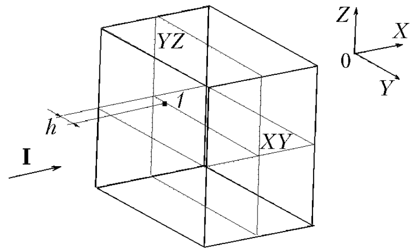


Fig. 3. Schematical representation of the location of the defect (1) in the posistor (the *YZ* plane is parallel to the electrodes).

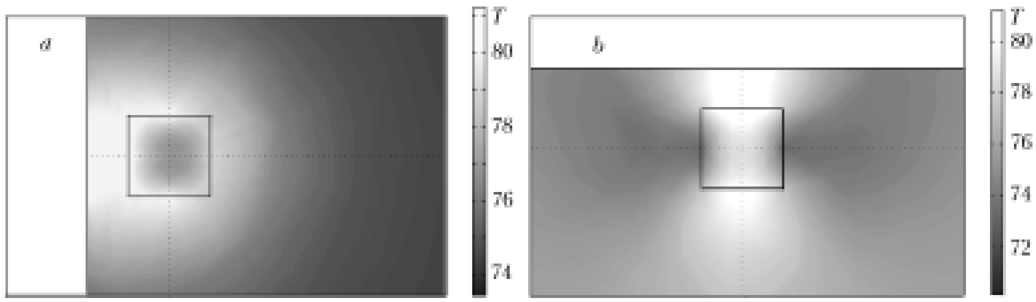


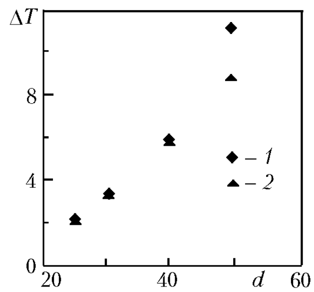
Fig. 4. Temperature distribution in the region of a pore of size 50 μm (after 2.5 msec) located at a distance of 50 μm from the side surface [(a) *YZ* plane, (b) *XY* plane].

TABLE 1. Time Change in the Maximum Temperature Differential ΔT_{max} in the Region of Voids of Different Sizes

d, μm	t, msec								
	1.0	1.5	2.0	2.5	3.0	3.5	4.0	5.0	10.0
25	1.96	2.35	2.34	2.08	1.73	1.38	1.04	0.61	0.2
30	2.86	3.45	3.12	3.12	2.59	2.04	1.52	0.92	0.4
40	4.56	5.64	5.35	5.35	4.56	3.82	3.03	1.83	0.6
50	6.17	7.90	8.59	8.21	7.29	6.37	5.15	3.21	1.0

perature differential is observed a few milliseconds after the application of the electric field and increases with increasing size of the defect. Upon switching on of the electric field the region near the defect has the highest temperature. At this site the material first reaches Curie temperature, i.e., in this region the specific resistance (PTCR effect) begins to increase sooner. Consequently, there occurs a redistribution of the electric field — an increase in the electric field strength in the region with a higher specific resistance. The heat generation in the ceramics in the region of the side surface of the void begins to grow markedly, which leads to an increase in the temperature differential. As soon as the main volume of the posistor reaches the temperature at which the resistance begins to increase, the local temperature differential sharply decreases and becomes insignificant, since the electric field in the region of the defect becomes more homogeneous and the current density on the whole markedly decreases. Thus, a local temperature burst takes place after the temperature at which the PTCR effect in the region of the defect is reached. With increasing size of the void ΔT_{max} is attained after a longer time upon the application of voltage.

For the case of inclusion of a large grain, the temperature field distribution pattern is opposite. The heat generation near the grain surfaces parallel to the posistor electrodes is maximal, and in the region of the side surfaces a local minimum of temperature is observed. It should be noted, however, that a void produces a much stronger effect on the temperature field distribution than a grain of the same size (as to the value of the temperature differential). Therefore, let us consider in more detail defects in the form of voids.



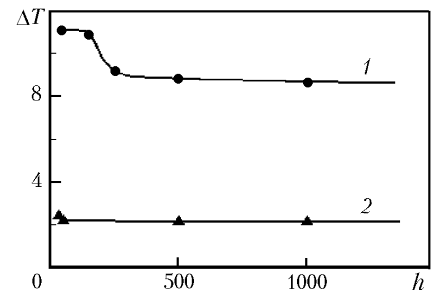


Fig. 5. Value of the temperature differential in the region of a pore (after 2.5 msec) located at a distance of 50 μm (1) and 500 μm from the side surface depending on its size.

Fig. 6. Temperature differential in the region of a void (after 2.5 msec) as a function of the distance from the side surface of the thermistor: 1) void of size 50 μm; 2) 25 μm.

The value of the temperature differential in the region of a pore linearly depends on both the size of the defect and its location. Let us emphasize that we considered the location of the defect in the central plane (*YZ*) parallel to the posistor electrodes. In this plane, delamination of the posistors occurs. Figure 5 shows the change in the temperature differential depending on the size of the pore and its distance from the side surface of the posistor.

In the case where the distance from the side surface of the posistor to the void considerably exceeds its size, the dependence of the temperature differential on the void size is well described by a polynomial of the 2nd power $\Delta T = ad^2 + bd$ (*a* and *b* are constants). If the pore size is no more than 10 μm, the local temperature differentials do not exceed 1^{o}C. The maximum influence on the temperature distribution is produced by the pores located at a distance from the side surface of the thermistor comparable to the size of the defect (Fig. 6).

From Figs. 5 and 6 it is seen that large inclusions of the air phase (> 50 μm) cause rather large temperature differentials whose sizes increase in the vicinity of the side surfaces of the thermistors.

For thermistors with a high specific resistance, the local temperature differentials are insignificant. For example, at a specific resistance of 5 Ω·m the temperature differential in the region of a void of size 50 μm is equal to a few tenths of a degree.

Conclusions. Defects in the form of voids strongly influence the temperature fields in the PTCR thermistors. For low-resistance semiconductor ceramics based on barium titanate, the temperature differentials in the region of voids of size 50 μm and more can reach dozens of degrees. The temperature gradients are most significant in the region of defects located near the side surfaces of the posistors. The maximum of the local temperature differential is observed after the material in the region of the defects reaches the temperature at which the PTCR effect begins.

NOTATION

c, specific heat capacity, J/(kg·K); *d*, thickness (size) of the defect, μm; **I**, electric current, A; *h*, distance of the defect to the side surface of the posistor, μm; *L*, posistor thickness, m; **n**, unit normal vector to the surface; *Q*, volume density of the thermal flux, $J/(m^3 \cdot sec)$; *T*, temperature, ^{o}C; T_0, environment temperature, ^{o}C; ΔT, temperature differential in the vicinity of the defect, ^{o}C; T_C, Curie temperature, ^{o}C; *t*, time, msec; λ, heat conductivity coefficient, W/(m·K); ρ, specific resistance, Ω·m; ρ^*, density, kg/m^3; σ, heat transfer coefficient of the surface, $W/(m^2 \cdot K)$; φ, electric field potential, V; ∇, Hamilton operator. Subscripts: max, maximum; 0, environment.

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