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MODELING OF LOCAL THERMAL FIELDS IN SEMICONDUCTING BARIUM TITANATE CERAMICS

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A mathematical model for describing local temperature fields in barium titanate ceramics with a positive temperature coefficient of resistance under the action of electric current has been constructed. The space-time temperature distribution within the grains of semiconducting barium titanate with varying microstructure and specific resistance has been studied.

Introduction. Ceramic materials based on barium titanate doped with rare-earth elements possess a unique property consisting of an abnormal increase in specific resistance — the effect of a positive temperature coefficient of resistance (PTCR). According to the Heywang model, the PTCR effect is conditioned by the formation of barrier layers at the grain boundaries at a temperature higher than that of phase transition [1]. Thermal resistors based on semiconductor $BaTiO_3$ have found wide application in electrical engineering and electronics as heating and temperature-sensitive elements, elements of protection against current overloads, and others [2]. In all of the cases listed, the material passes repeatedly through the Curie temperature. Barium titanate possesses a first-order ferroelectric phase transition. In the region of Curie temperature some of the characteristics of this material change significantly: there occurs a jump in heat capacity and permittivity and the coefficient of thermal linear expansion reverses sign and becomes negative [3]. On heating a thermal resistor by electric current, significant thermal gradients are observed in samples, i.e., not the whole of the material is involved in phase transition. In a number of cases (in circuits of protection against high current overloads and under conditions of intense heat exchange with the environment), the temperature gradients become responsible for the appearance of large thermoelastic stresses commensurable with critical voltages which lead to the appearance and propagation of cracks and sharp deterioration of the electric characteristics of samples [4, 5]. Two components of the temperature stresses can be singled out: 1) parabolic temperature distribution through the thickness of samples which does not take into account the local structure of ceramics; 2) local temperature inhomogeneities within one grain owing their origin to the characteristic features of the electrophysical parameters of semiconducting barium titanate ceramics, since the grain has a high-resistance boundary layer and low-resistance interior. According to [6], the nonuniformity of the thermal field within the grain–boundary system 50 μm in size may reach several tens of degrees, which is commensurable with a temperature drop over a sample of thickness equal to several millimeters without allowance for its granular structure. It should be noted that contemporary technological processes of production allow one to obtain a sufficiently homogeneous ceramics with the size of a grain not exceeding 10 μm. Local temperature gradients are determined not only by the grain sizes but also by the properties of the material (specific resistance, positive temperature coefficient of resistance, and phase-transition temperature). Of particular interest is a detailed study of the dynamics of a change in the distribution of local thermal fields in the process of switching of thermal resistors to a high-resistance state (at transition through the Curie temperature). The purpose of the present study is to investigate the temperature distribution over the grains of semiconducting barium titanate of different microstructures and at different characteristics of the ceramics.

Theoretical Model. In the general case, ceramics represents a complex parallel–series connection of resistors that imitate "grains" and "boundaries." In the simplest case, one may confine oneself to consideration of one consecutive chain of identical grains. Then the distribution of a thermal field in one grain may be described by solving the one-dimensional heat-conduction equation

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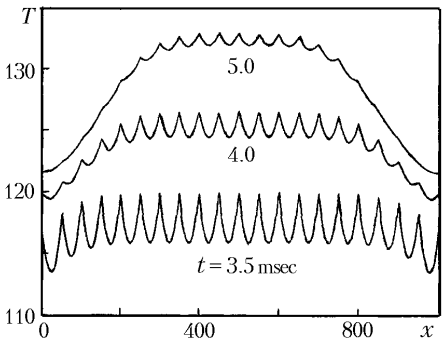


Fig. 1. Temperature distribution in the thermal resistor for different time moments (T_C = 120°C, ρ = 0.4 Ω·m, d = 50 μm).

$$c\rho^* \frac{\partial T}{\partial t} - \lambda \Delta T = q \,. \tag{1}$$

This equation must obey the following initial condition: $T(x, t) = T_0$ at $t = 0$.

The boundary conditions were selected proceeding from the assumption that the heat flux on the surface was proportional to the difference in the temperatures of the surface and the environment (Newton's law):

$$\left.\frac{\partial T}{\partial x}\right|_{x=0} + \sigma\,(T(0, t) - T_0)/\lambda = 0 \,, \tag{2}$$

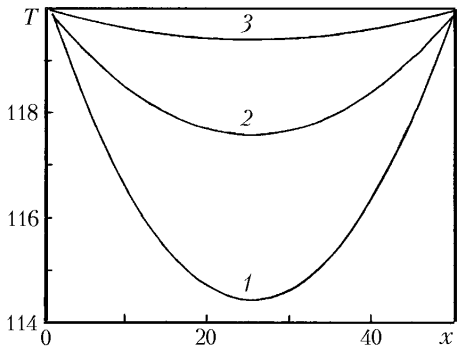
$$\left.\frac{\partial T}{\partial x}\right|_{x=d} - \sigma\,(T(d, t) - T_0)/\lambda = 0 \,. \tag{3}$$

The bulk density of the heat flux in Eq. (1) depends on the temperature of a volume element $q = q(T)$. Thus, we have a nonstationary boundary-value problem with nonlinearity of the third kind [7]. In the case where the specific resistance ρ depends on temperature, the following relation holds:

$$q(x, T) = \frac{U^2 \rho(x, T)}{\left(\int_0^d \rho(x, T)\, dx\right)^2} \,. \tag{4}$$

It should be noted that the specific resistance of the grain boundary differs from that in the interior of the grain. Here the PTCR effect owes its origin to the change in the barrier boundary layer. The boundary thickness is an order of magnitude smaller than the grain size [8]. With consideration for this fact and also for the data obtained from Kole–Kole's diagrams [9], it can be assumed that the difference between the specific resistances of the boundary and volume of the grain amounts to 2–3 orders of magnitude in a low-resistance state (lower than T_C). The specific heat of posistor ceramics ρ depends on the material composition, technological regimes of its manufacturing, the temperature of the sample, and the voltage applied (varistor effect). It is difficult to approximate the temperature dependence of the specific resistance of the grain boundary by one mathematical expression within the whole range of working temperatures. A generalized or "typified" temperature characteristic: which is noted for simplicity and provides a correct idea of the character of the change in the resistance of the material studied was used

$$\rho(T) = \begin{cases} \rho_0 \,, & T < T_C \,; \\ \rho_0 \exp\left\{\alpha\,(T - T_C)\right\}, & T_C \le T < T_{\max} \,; \\ \rho_0 \exp\left\{\alpha\,(T_{\max} - T_C)\right\}, & T > T_{\max} \,, \end{cases} \tag{5}$$



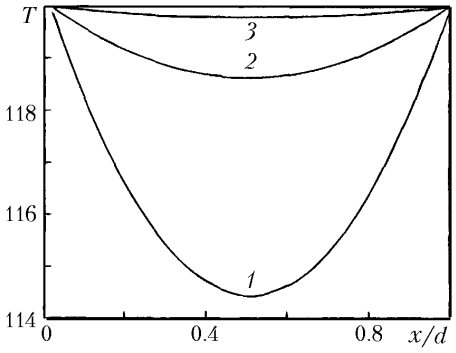


Fig. 2. Temperature distribution over the grain at different specific resistance values (T_C = 120°C, d = 50 μm): 1) ρ = 0.4; 2) 1.0; 3) 4.2 Ω·m.

Fig. 3. Temperature distribution at different sizes of grains (T_C = 120°C, ρ = 0.4 Ω·m): 1) d = 50; 2) 25; 3) 10 μm.

TABLE 1. Minimum Temperature Drop in the Grain (T_C = 120°C, E = 220 W/mm)

ρ, Ω·m	d, μm		
	10	25	50
0.42	0.219	1.368	5.45
1	0.0913	0.570	2.28
4.2	0.0219	0.1370	0.548

where $\alpha(x)$ is the PTCR coefficient.

The grain was broken into 50 layers, two of which were considered as a boundary, with the dependence of the specific resistance on temperature according to (5). The difference between the specific resistances of the boundary and of the grain was taken equal to 500 (at a temperature lower than T_C). Equation (1) was solved by an implicit numerical method (Crank–Nicholson scheme) [10] and an explicit one (Dufour–Frankel scheme). Both methods gave close results.

Results and Discussion. Figure 1 presents the results of calculations of temperature fields in large-grain ceramics at different instants of time after application of an electric field of 200 W/mm to a sample (a chain of 20 grains, each of size 50 μm, was considered). In the process of ceramics heating, significant temperature gradients are observed within a grain. This is due to the generation of a large amount of heat in the boundary layer that has a higher resistance. A maximum temperature gradient is observed when the boundary temperature reaches the phase-transition temperature. In a steady state, the temperature drop in the grain becomes insignificant. The data for the grains located in the central part of samples agrees well with the results obtained for a thermally insulated grain [6]. Therefore, in what follows we may limit ourselves to consideration of thermal fields in a thermally insulated grain.

Figures 2 and 3 show changes in temperature within one grain at different values of the specific resistance and size of the grain, respectively. As far as a thermally insulated grain was considered, for boundary conditions (2) and (3) the heat-transfer coefficient was assumed equal to zero. The value of the temperature gradient increases with a decrease in the specific resistance and increase in the grain size. As is seen from Table 1, on decrease in the specific resistance by a factor of 10, the value of ΔT increases practically the same number of times. At the same time, on increase in the grain size by a factor of 5, the value of ΔT increases 20 times, i.e., the grain size exerts a predominant influence on the maximum temperature gradient in a thermally insulated grain.

As was noted, the difference between the specific resistances of the boundary and grain was considered equal to 500 (at a temperature lower than T_C.) Therefore, heat generates on the thin boundary of the crystallite. The entire grain is heated by the heat flux outgoing from the boundary. On attainment of the maximum temperature difference, the value of ΔT remains virtually intact till T_C is reached. When the grain boundary temperature exceeds T_C, its resistance begins to increase exponentially with a respective decrease in heat generation.

Note that the temperature drop over the grain depends significantly on the voltage applied to the sample. The dependence of ΔT on the electric-field intensity is close to a quadratic one, since heat generation is proportional to U^2 (Eq. (4)).

Conclusions. Based on numerical solution of the heat-conduction equation, the space–time distribution of temperature in the grains of semiconducting barium titanate ceramics has been studied. It is shown that the temperature difference between the interior and the boundary of the grain of ceramics increases with a decrease in the specific resistance and increase in the grain size. The temperature gradient over the grain may exceed 5^{o}C for large-grain and low-resistance ceramics. In the case of a small-grain structure (grain size less than 10 μm), the value of ΔT is insignificant. The dependence of ΔT on the electric-field intensity obeys the quadratic law.

NOTATION

c, specific heat, J/(kg·K); d, thickness, μm; E, external electric-field intensity, W/mm; q, bulk density of heat flux, J/(m^3·sec); T, temperature, ^{o}C; T_C, Curie temperature, ^{o}C; T_{max}, maximum thermistor temperature, ^{o}C; T_0, environmental temperature, ^{o}C; t, time, msec; U, voltage applied to the posistor, V; x, coordinate, μm; α, temperature coefficient of resistance, %/K; λ, thermal conductivity, W/(m·K); ρ, specific resistance, Ω·m; ρ^*, density, kg/m^3; σ, heat-transfer coefficient of the surface, W/(μ^2·K); Subscripts: max, maximum; 0, environment.

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