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TEMPERATURE STRESSES IN BARIUM-TITANATE-BASED SEMICONDUCTOR CERAMICS

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Temperature stresses in barium-titanate-based semiconductor ceramics in the process of heating by an electric current have been investigated by numerical methods. The fracture of low-resistance thermistors by the delamination mechanism has been substantiated.

Introduction. Barium-titanate-based semiconductor ceramics possesses the property of an anomalous increase in the specific resistance (resistivity) above the Curie temperature (T_C). This phenomenon is known as a positive temperature coefficient of resistance (PTC). According to the Heywang model, this effect is related to the formation of barrier layers at the grain boundaries above the Curie temperature [1]. Thermistors of the PTC type (posistors) based on barium-titanate semiconductor ceramics are widely used as protection devices, heating elements, and temperature sensors [2]. In the case of the action of heavy current loads, mechanical failure of the thermistors occurs, which is caused by the significant temperature gradients in samples. We know of three basic types of mechanical failure [3–5]: formation of an irregular network of cracks perpendicular to the basic surfaces, formation of cracks on the lateral disk surface or at the periphery of the electrodes, and failure by propagation of the crack on a plane that is in parallel to the electrodes, when the posistor is split into two equal halves. The first two types are related to the inhomogeneities and defects of the ceramics and are dependent, as a rule, on the quality of production of the samples. The reason for the last type of failure (it is called the "delamination" effect) is more fundamental: it is the fracture of the ceramics.

Theoretical and experimental investigations of the temperature fields in posistor elements have been performed in [5–8]. In particular, it has been shown that the temperature difference between the center and surface of a posistor can attain 20–40^{o}C. Temperature stresses have been described most completely in [5], where the maximum tensile stresses in a homogeneous ceramics were shown to be ~15 MPa. The limiting tensile stresses for posistor ceramics have a value of 50–100 MPa [9, 10]. In this connection, a more detailed study of the temperature stresses produced in thermistors by the action of heavy current loads is necessary for explaining the delamination effect. This work seeks to investigate temperature stresses in posistor elements in the process of heating by an electric current.

Theoretical Model. For the considered dimensions of posistor elements, displacements as a result of thermal expansion are insignificant; therefore, we can disregard inertia forces and restrict our consideration to the quasistatic thermoelasticity problem. The influence of strain as a result of thermal expansion on the temperature field is also slight [11]. Since protective posistors have, as a rule, the shape of cylinders to whose bases electrodes are applied, it is convenient to select a cylindrical coordinate system (z, r, φ). Because of the symmetry of the element (the properties are independent of the coordinate φ and are symmetric about the plane that is parallel to the electrodes and passes through the center ($z = 0$), we can restrict ourselves to an analysis of the case of axial symmetry. It is sufficient to consider half the posistor, since its second half can be obtained by mirror reflection. Mechanical stresses in elasticity theory are described for a three-dimensional space using the second-rank tensor consisting of six components. In the case of axial symmetry we will have only the following nonzero components: τ_r, τ_z, $\tau_{rz} = \tau_{zr}$, and τ_φ. To determine the stress tensor (and four nonzero components of the strain tensor ε_r, ε_z, ε_{rz}, and ε_φ and two components of the displacement vector u_r and u_z) we must solve a system of ten equations [11–13].

The first two equations (static equations) can be obtained from the equilibrium condition

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$$\frac{\partial \tau_r}{\partial r} + \frac{\partial \tau_{rz}}{\partial z} + \frac{\tau_r - \tau_\varphi}{r} - F_r = 0 , \tag{1}$$

$$\frac{\partial \tau_z}{\partial z} + \frac{\partial \tau_{rz}}{\partial r} + \frac{\tau_{rz}}{r} - F_z = 0 . \tag{2}$$

Hooke's law enables us to obtain four more equations (physical equations)

$$\tau_z = \lambda \, (\varepsilon_z + \varepsilon_r + \varepsilon_\varphi) + 2\mu\varepsilon_z , \tag{3}$$

$$\tau_r = \lambda \, (\varepsilon_z + \varepsilon_r + \varepsilon_\varphi) + 2\mu\varepsilon_r , \tag{4}$$

$$\tau_\varphi = \lambda \, (\varepsilon_z + \varepsilon_r + \varepsilon_\varphi) + 2\mu \, \frac{u_r}{r} , \tag{5}$$

$$\tau_{rz} = \mu\varepsilon_{rz} . \tag{6}$$

The strain-tensor and displacement-vector components are related by the Cauchy formulas (geometric equations)

$$\varepsilon_z = \frac{\partial u_z}{\partial z} , \tag{7}$$

$$\varepsilon_r = \frac{\partial u_r}{\partial r} , \tag{8}$$

$$\varepsilon_\varphi = \frac{u_r}{r} , \tag{9}$$

$$\varepsilon_{rz} = \frac{\partial u_z}{\partial r} + \frac{\partial u_r}{\partial z} . \tag{10}$$

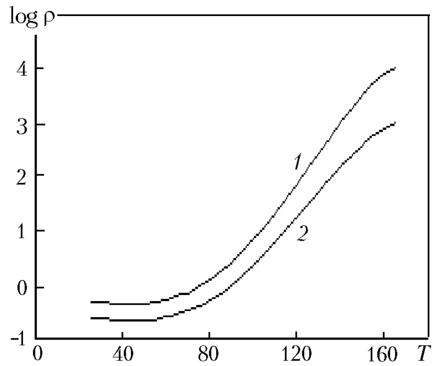
With allowance for the influence of temperature on mechanical stresses as a result of the thermal expansion (compression) of ceramics, Eqs. (3)–(5) will take the form

$$\tau_z = \lambda \, (\varepsilon_z + \varepsilon_r + \varepsilon_\varphi) + 2\mu\varepsilon_z - (3\lambda + 2\mu) \, \varepsilon^T , \tag{11}$$

$$\tau_r = \lambda \, (\varepsilon_z + \varepsilon_r + \varepsilon_\varphi) + 2\mu\varepsilon_r - (3\lambda + 2\mu) \, \varepsilon^T , \tag{12}$$

$$\tau_\varphi = \lambda \, (\varepsilon_z + \varepsilon_r + \varepsilon_\varphi) + 2\mu \, \frac{u_r}{r} - 3\lambda\varepsilon^T . \tag{13}$$

If the temperature dependence of the elongation per unit length does not obey the linear law (which is true of barium-titanate ceramics in the region of phase transition [3]), we must use the following expression [13]:



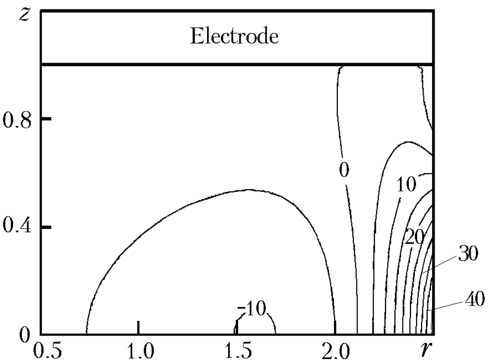


Fig. 1. Temperature dependence of the specific resistance of protective posistor ceramics: 1) zero electric-field strength; 2) electric-field strength 100 W/mm. T, ^{o}C; ρ, Ω·m.

Fig. 2. Distribution of the temperature stresses τ_z in the sample. Numbers at the curves, τ_z values, MPa. r, z, mm.

$$\varepsilon^T = \int_{T_0}^{T} \alpha (T)\, dT \,. \tag{14}$$

The temperature distributions for posistor elements based on barium-titanate semiconductor ceramics were obtained earlier by solution of the heat-conduction equations [7, 8]. Force boundary conditions were selected from the assumption that the posistor-element surface is unaffected by external forces:

$$\tau_r (z, D/2, t) = f_r = 0 \,, \tag{15}$$

$$\tau_z (h/2, r, t) = f_z = 0 \,. \tag{16}$$

By virtue of the symmetry of the problem in question, we specified the following kinematic conditions:

for the axis of symmetry $r = 0$

$$u_r (z, 0, t) = 0 \,; \tag{17}$$

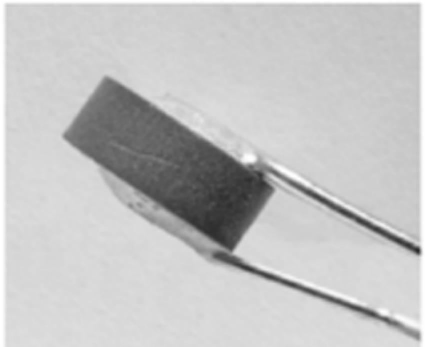
for the plane of symmetry $z = 0$

$$u_z (0, r, t) = 0 \,. \tag{18}$$

Thus, Eqs. (1), (2), and (6)–(18) enable us to determine mechanical stresses in the material; however it is difficult to find the analytical solution of this system. Therefore, we used the finite-element method (FEMLAB system) for the case of barium-titanate-based posistor ceramics, when the coefficient of thermal expansion is a function of the temperature in the region of phase-transition temperature.

Results and Discussion. To calculate temperature stresses we measured the characteristics of protective posistor elements manufactured by the "Monolit" Republic Unitary Enterprise. Figure 1 gives the temperature dependences of the specific resistance for a thermistor with switching temperature T_C = 87^{o}C and a specific resistance ρ_{25} = 0.31 Ω·m. It is seen that the varistor effect (change in the specific resistance with electric-field strength) has a significant influence on the value of the specific resistance and consequently on thermophysical processes. Therefore, the temperature dependences of the specific resistance were approximated for modeling with allowance for this effect.

It is common knowledge that the tensile mechanical strength of ferroelectric ceramics is virtually an order of magnitude lower than the compressive strength [9]. Therefore, tensile stresses are the reason for the failure of ceramic



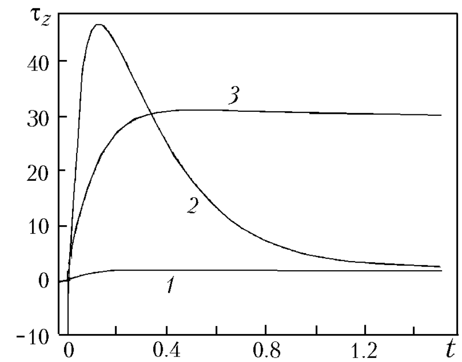


Fig. 3. Photograph of the lateral surface with a crack for the posistor after a few connection–disconnection cycles.

Fig. 4. Temperature stresses vs. time at the center of the lateral thermistor surface: 1) for the posistor without an electrode, σ = 50 W/(m^2·K); 2) for the posistor with an electrode, σ = 50 W/(m^2·K); 3) for the posistor without an electrode, σ = 800 W/(m^2·K). t, sec; τ_z, MPa.

TABLE 1. Maximum Temperature Stresses τ_z (MPa) of Posistors with a Switching Temperature of 87°C

ρ_{25}, Ω·m	d, μm							
	0	25	50	100	150	200	250	300
0.62	1.86	20.1	27.6	35.4	40.6	44.4	47.4	52.8
0.31	1.90	22.9	29.4	37.1	44.0	46.7	50.0	54.6
0.16	1.95	24.4	30.8	38.5	47.8	49.9	52.3	58.6

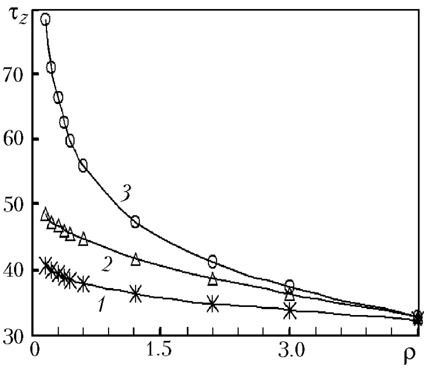
samples as a rule. Figure 2 shows the distribution of the mechanical stresses τ_z for the instant of time t = 0.13 sec, when the tensile stresses are maximum. It is seen that the maximum tensile stresses are concentrated at the center of the lateral thermistor surface, and their value can attain 50 MPa. The critical tensile stresses for barium-titanate-based semiconductor ceramics are 50–100 MPa [9, 10], which is comparable with the calculated values.

Thermistors fail, as a rule, by the delamination mechanism in a few connection–disconnection cycles. Growth in the resistance at room temperature is recorded after each cycle. The formation of cracks on the lateral surface is observed (Fig. 3), which is consistent with calculation results.

The value of the tensile stresses τ_z is dependent on a number of factors: the electrode thickness, the intensity of heat exchange with the environment (coefficient of heat transfer of the surface), and the geometry and properties of the ceramics (switching temperature T_C and specific resistance ρ). Figure 4 gives the values of the stresses τ_z at the center of the lateral surface at different instants of time after the application of voltage for thermistors with tin-lead electrodes of thickness d = 200 μm and without them for different heat-transfer coefficients. In the latter case the presence of a thin-film current-conducting layer with a thickness of the order of 1 μm, which exerts no influence on the temperature distribution, is meant.

The stress jump in the first tens of milliseconds is related to the change of sign by the coefficient of thermal expansion of barium titanate in phase transition and does not exceed −10 MPa. In the absence of tin-lead electrodes, the value of tensile stresses is substantially dependent on the heat-transfer coefficient and virtually remains constant, once the maximum has been obtained. Even for high values of the coefficient of heat transfer of the surface σ = 800 W/(m^2·K) (which corresponds to the case of operation of thermistors as part of thermal fans), we have $\tau_z \sim$ 30 MPa.

The tensile stresses τ_z in posistors with electrodes attain their maximum in a temperature interval of 100–200 msec after the application of electric voltage. This is due to the intense drawing of heat from the ceramics to the electrode, which contributes to the formation of a high temperature gradient. The temperature field, in the stationary state, is equalized across the thickness after the heating of a fairly thick electrode, which results in a decrease in the tensile stresses. The maximum tensile stresses can exceed 50 MPa for an electrode thickness greater than 200 μm (see Table 1).



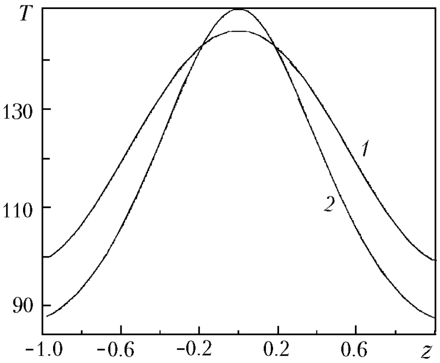


Fig. 5. Temperature stresses vs. specific resistance: 1) for the posistor without an electrode, σ = 1000 W/(m^2·K); 2) for the posistor with an electrode, σ = 50 W/(m^2·K); 3) for the posistor with an electrode and with a multiplyer resistor, σ = 50 W/(m^2·K). ρ, Ω; τ_z, MPa.

Fig. 6. Temperature distribution across the thickness of a posistor with electrodes at the instant of attainment of the maximum tensile stresses (D = 5 mm and d = 200 μm): 1) without a ballast resistor (t = 0.13 sec); 2) with a ballast resistor of 90 Ω (t = 0.2 sec). T, ^{o}C.

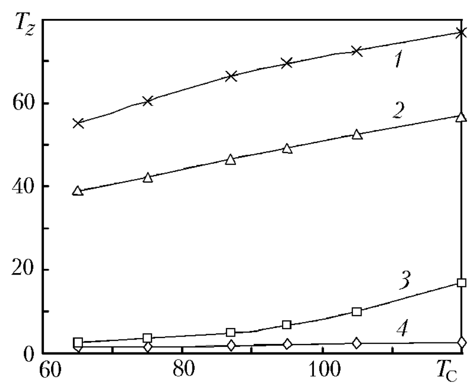


Fig. 7. Temperature stresses vs. switching temperature (σ = 50 W/(m^2·K): 1) for the posistor with an electrode and a multiplyer resistor; 2) for the posistor with an electrode and without a multiplyer resistor; 3) for the posistor without an electrode and with a multiplyer resistor. T_z and T_C, ^{o}C.

The value of the maximum tensile stress τ_z is also determined by the properties of semiconductor ceramics (switching temperature and the specific resistance). The stresses decrease as the specific resistance increases (Fig. 5).

Thermistors of the PTC type are usually the elements of protection against current and power overloads. Therefore, they are series-connected to electric circuits with a certain load resistor. A ballast resistor (R = 90 Ω) limiting the current to 2 A is analogously connected to the electric circuit in series with the protective posistor. The presence of the limiting resistance contributes to the increase in the temperature stresses (Fig. 5, curve 3). The effect of the multiplyer resistance is as follows: when the current in the circuit is limited, the process of heating of the thermistor slows down, which results in the increased inhomogeneity of the temperature field due to the outflow of heat from deeper electrode regions of the ceramics (fig. 6). The higher inhomogeneity of the temperature field leads to higher values of the temperature stresses.

The temperature difference across the sample's thickness increases with switching temperature [7], which causes the maximum tensile stresses to grow (Fig. 7).

Conclusions. We have studied temperature stresses in PTC-type thermistors in the process of heating by an electric current. It has been established that the maximum tensile stresses are concentrated at the center of the lateral thermistor surface, and their values grow with decrease in the specific resistance and increase in the switching temperature. In the presence of tin-lead electrodes (of thickness 200 μm or more), the tensile stresses can exceed 50 MPa, which is comparable with the critical values for barium-titanate-based semiconductor ceramics. Thus, on the basis of the numerical experiments, we have explained why thermistors fail by the delamination mechanism.

NOTATION

D, posistor diameter, mm; *d*, electrode thickness, μm; *F*, body force, N/m^3; *f*, surface force, N/m^2; *h*, posistor thickness, mm; *R*, ballast resistance, Ω; *r*, *z*, coordinates, mm; *T*, temperature, ^{o}C; T_0, temperature at which the thermal expansion is taken to be zero, ^{o}C; *t*, time, sec; *u*, displacement, m; α, coefficient of thermal expansion, K^{-1}; ε, elongation per unit length; λ and μ, Lamé constants, Pa; ν, Poisson coefficient; ρ, specific resistance, Ω·m; σ, coefficient of heat transfer of the surface, $W/(m^2 \cdot K)$; τ, temperature stress, MPa. Subscript: 25, at a temperature of $25^{o}C$.

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