THERMOPHYSICAL PROPERTIES OF ELECTRICAL INSULATION AFTER MULTIPLE COOLING AND IRRADIATION AT TEMPERATURES OF 4.2-400 K

S. A. Tanaeva and L. E. Evseeva

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Thermophysical properties of electrical insulation in cryogenic power devices are investigated experimentally over a wide temperature range.

Polyimide glass-reinforced plastics are a promising electrical insulation for various high-power installations operating at cryogenic temperatures. During use the electrical insulation of these devices is under the joint action of large mechanical loads of electromagnetic origin, extremely low temperatures, irradiation, and pulse or constant electric fields. Disruption of the integrity or a drastic deterioration of the properties of the insulation under any of these actions is inadmissible. Therefore the following demands are made on it [1]: electrical strength (it must be not lower than 500 V at 4.2 K); high mechanical characteristics; high radiation resistance (the material must withstand a total fluence of $10^8 - 10^9$ rad); cryogenic resistance (the properties must not deteriorate in multiple coolings down to cryogenic temperatures); relatively high values of thermal conductivity and specific heat at low temperatures.

The present work sought to carry out a complex investigation of the influence of thermal cycling and irradiation on the thermophysical properties of two types of electroinsulating materials: materials reinforced with a polyimide glass cloth and impregnated with an epoxyphenol compound (LSNL) and those reinforced with a polyimide film and a polyimide varnished glass cloth (alternating) and impregnated with the same compound (LSNL+PML). The polyimide film and polyimide varnish serve to impart higher radiation resistance to the materials.

The thermophysical properties (the coefficients of thermal conductivity λ and thermal diffusivity a and the specific heat C) of these materials are investigated experimentally over a wide temperature range (4.2-400 K); a quasistationary method of determination of λ , a, and C in a complex as a result of one experiment is used [2]. The limiting relative error is 7-8%.

Besides, since the given materials have a moderately high porosity, their pycnometric density and porosity were determined in the initial state and after thermal cycling (77-373 K) on a Micromeritics helium pycnometer. The LSNL material porosity is 22.4% and is almost unchangeable in thermal cycling. The hybrid LSNL+PML material porosity is equal to 14.3% and increases in thermal cycling according to the linear law

$$\Pi = 14.3 + 0.035n \, ,$$

where n is the number of cycles of the thermal shocks.

For the LSNL material this formula has the form

$$\Pi = 22.4 + 0.01n \, .$$

The density in thermal cycling decreases for the first five cycles and then stabilizes.

Cryogenic Resistance. Figure 1 gives the temperature dependences of the thermal conductivity coefficient of the investigated materials in the initial state and in thermal cycling. There is a "plateau" at low temperatures,

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Fig. 1. Thermal conductivity of the LSNL and LSNL+PML materials vs. number of thermal cycles: 1) the initial state, 2) n = 1, 3, 5, 4) 20. λ , W·m⁻¹; *T*, K.



Fig. 2. Change in the thermal conductivity and specific heat of the LSNL+PML (a) and LSNL (b) material before (λ_0, C_0) and after (λ_n, C_n) thermal cycling: 1) n = 1, 2 5, 3) 20.

characteristic of amorphous polymers and glass-reinforced plastics on their base [3]. For the LSNL material it is in the interval 8–15 K and for the LSNL+PML material it is from 6 to 11 K, which is associated with a smaller percentage of glass in the LSNL+PML material (35.6%) than in the LSNL material (53.9%). The absolute value of $\lambda_{\text{LSNL+PML}}$ is much higher than that for the LSNL material, which correlates with their porosity. For this reason we observe a substantial dependence of λ on the type and pressure of the heat-transfer gas. Thus, for the LSNL



Fig. 3. Values of the coefficients C and a in the initial state. a, $m^2 \cdot \sec^{-1}$; C, $kJ \cdot kg^{-1} \cdot K^{-1}$.

material the ratio of λ in the helium medium at P = 1 atm to λ in the helium medium at $P = 10^{-2}$ mm Hg (in vacuum) changes in the temperature range under study from 1.5 to 2.1. For the hybrid material this ratio is 1.25.

Multiple cooling variously affects λ of these materials. Whereas after the first cooling cycle λ of both materials increases somewhat (Fig. 2), subsequent thermal cycling (up to 5) leads to a decrease in $\lambda_{\text{LSNL+PML}}$, while λ_{LSNL} increases further. Figure 3 gives the temperature dependences of the specific heat C and thermal diffusivity a of the given materials in the initial state. In absolute value $C_{\text{LSNL}} < C_{\text{LSNL+PML}}$, since the percentage of glass is larger in the LSNL material. A series of relaxation transitions is observed on C(T); multiple cooling, having practically no effect on the absolute value of C (the exception is only the region T < 15 K) (Fig. 2), displaces the temperatures of these relaxation transitions into a lower region and increases the amplitude of the change in C at the given temperature.

Analysis of the results obtained indicates that in the LSNL material in thermal cycling the processes of cross-linking and rehardening are dominant (changes in λ and *C* proceed oppositely at T < 15 K) and in the LSNL+PML material cross-linking is possible only in the first cycles of thermal shocks and thereafter formation of microcraks occurs on the film-binder interface since a high surface energy of the polyimide film makes it impossible to create a strong adhesion bond between the film and the binder.

Radiation Resistance. The interaction of radiation with organic materials is accompanied by radiation destruction and radiation cross-linking. The chemical structure strongly affects the direction of basic radiation-chemical processes in organic materials: either prevailing cross-linking or prevailing destruction occurs. Radiation cross-linking is observed as a result of formation of chemical bonds between macromolecules or different portions of one macromolecule; a reticular structure forms. Radiation destruction is the processes of disruption of the main polymer chain, decomposition of the spatial network nodes, separation of the lateral groups [4], etc.

In organic materials both processes proceed simultaneously but with different intensity. Thus, phenolformaldehyde and epoxide resins and polyimides predominantly cross-link in irradiation [3]. The interaction of radiation with organic materials is accompanied by gas release. Analyzing the gaseous products of radiolysis of phenolformaldehyde and epoxide resins showed that they consist mainly of two-element gases (H_2 , CO, CO₂, CH₃, and other hydrocarbons) [3]. Gas release decreases thermal conductivity due to the formation of cracks and microdefects. Radiation destruction also decreases thermal conductivity and increases heat capacity; radiation cross-linking, on the contrary, increases thermal conductivity and decreases heat capacity.

Irradiation was performed by γ -quanta from a ⁶⁰Co source with an energy of 1.4 MeV. The total fluence was 10⁸ rad.



Fig. 4. Change in the thermal conductivity and specific heat before (λ_0, C_0) and after (λ_{rad}, C_{rad}) irradiation: 1) LSNL (1 day, 14 days), 2) LSNL+PML (1 day), 3) LSNL+PML (2 days).

Figure 4 gives the temperature dependences of the relative changes in thermal conductivity and specific heat before and after irradiation. A substantial postradiation change in $\lambda_{LSNL+PML}$ is recorded. Thus, λ , when two days have elapsed after irradiation, is found to be 10-15% higher than λ after the first day. It is obvious that with time once the irradiation has ceased the defects "heal" and the bonds restore. For the LSNL material, the absolute values of λ before and after irradiation (after a day or 14 days) practically coincide over the entire temperature range. Hence it follows that it is the bonds on the film-binder interface that are most subject to the action of irradiation. The absolute values of heat capacity before and after irradiation coincide for both materials over practically the entire temperature range, except for the low temperature region, with *C* decreasing in thermal cycling and increasing in irradiation (~80%). It can be assumed that since the heat capacity at the given temperatures is determined only by the vibrational spectrum of the lattice, the irradiation affects precisely the chemical intramolecular structure by "pulling out", for example, nitrogen and oxygen atoms and elementary hydrocarbons, thus creating additional defects (cavities) and leading to an increase in the specific heat [5]. With an increase in temperature, when the intermolecular interaction becomes decisive, the formation of the reticular structure due to radiation cross-linking leads to a decrease in the heat capacity to the level of the specific heat of nonirradiated material.

In conclusion it should be noted that at low temperatures the most sensitive characteristic for investigating radiation and cryogenic resistance is the specific heat while at high temperatures it is the thermal conductivity.

From the viewpoint of practical applicability the LSNL material is more suitable than LSNL+PML for use at low temperatures under irradiation.

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