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Structural Parameter for Estimating Durability of Composite Materials with Polymer Components in Strong Electric Fields

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Abstract. The purpose of the present work is to reveal and to substantiate the structural parameter for estimation of the durability of composite materials with polymeric components in strong electric fields. Rubbers filled with technical carbon are chosen as objects of research. They possess almost zero water absorption, resistance to aggressive media, high electrical and mechanical strengths, capability of acquiring practically any form, etc. Because of this, they are widely used in high-voltage electrophysics and electrical engineering. Composite materials are represented by an equivalent circuit of a series-parallel connection of a set of individual contacts conductive phase–dielectric–conductive phase formed by an electroconductive grid. The processes in the materials are considered on three hierarchical levels: individual contact conductive phase–dielectric–conductive phase, distribution of the set of individual contacts over their properties taking into account the dispersion of their parameters depending on the employed initial components, macrostructural level—the electroconductive grid imbedded into a polymer matrix. As a result of our investigations, the structural parameter—the entropy of the textural parameter that can be used to estimate the durability of composite materials with polymer components in strong electric fields—has been chosen.

Keywords: structural parameter, electric field strength, non-uniform structure, conductive phase, agglomeration

INTRODUCTION

At present electric systems operate under conditions of enhanced voltage levels and high current loads. In electrophysics the application of devices operating with high-power high-voltage pulses (generators of pulsed voltage, super high-power laser systems, etc.) is increased. Under these more stringent operating conditions, the role of composite materials that can operate in electric fields having extremely high strengths—prebreakdown electric voltages—also increases [1–3].

In composite materials the electrothermal breakdown mechanism is typically observed; therefore, an increase in the prebreakdown electric voltage is typically reduced to the development of a multicomponent structure with electric field intensity distribution over the material volume as uniform as possible. This calls for a study of physical principles of structurization and formation of properties determining the behavior of a composite material in strong electric fields.

The purpose of the work is to reveal and to substantiate the structural parameter for estimation of the durability of composite materials with polymer components in strong electric fields. An equivalent circuit of resistive composite material is a series-parallel connection of a set of individual contacts conductive phase–dielectric–conductive phase formed by an electroconductive grid [4]. Therefore, the voltage withstand without breakdown depends on the electric field strength in individual contacts.
OBJECT AND METHOD OF INVESTIGATION

The multicomponent material is represented as a hierarchical structure. The processes are considered on different levels:
– individual contact conductive phase–dielectric–conductive phase;
– distribution of the set of individual contacts over their properties taking into account the dispersion of their parameters depending on the initial components;
– macrostructural level—the electroconductive grid imbedded into a polymer matrix.

Rubbers filled with technical carbon were chosen as objects of research. They possess almost zero water absorption, resistance to aggressive media, high mechanical durability, capability of acquiring practically any form, etc. [5].

The calculation model described in [6] was used to study interrelation between the voltage per individual contact and the factors caused by adjustment of initial components.

RESULTS

It is established that the electric field strength in an individual contact depends on the work function of an electroconductive particle and thickness of the dielectric layer between electroconductive particles. With its increase, the influence of the work function weakens. Among the processes determining the dielectric layer thickness in the individual contact of the non-uniform structure is the agglomeration–deagglomeration of the filler in the polymer.

As is well known, the interlayer thickness of the individual contact depends on the concentration of the electroconductive filler in the composite material [7]. A decrease in the filler concentration increased not only the layer thickness, but also the non-uniformity of filler distribution over the matrix and hence the dispersion of sizes of the interlayer between conductive particles. Such non-uniformity causes a local increase in the electric field strength leading to breakdowns in strong electric fields. This reduces the maximum withstand voltage.

The factors that determine the field strength in the individual contact for a preset voltage in the design and the dispersion of individual contact parameters were studied in [8] using a probabilistic-deterministic model of agglomerated filler distribution over the matrix volume. The developed approach was based on the application of the packing factor of filler particles as a parameter that allows the polymer properties to be considered. To analyze conditions of occurrence and increase in the dispersion of sizes of dielectric interlayers in the individual contacts, the probability density function for the deviation of distances between the particles from their average value was determined:

\[
g(h) = \frac{3\sqrt{D^3}}{(\bar{h} + D)^3} \frac{1}{\sigma_c \sqrt{2\pi}} e^{-\frac{(\sqrt{D^3} - C_0)^2}{(2\sigma_c^2)}} \tag{1}\]

where \(\nu\) is the packing factor for filler particles, \(\bar{h}\) is the average distance between the particles, \(D\) is the average size of agglomerated particles, \(C_0\) is the filler concentration, and \(\sigma_c\) characterizes the dispersion of the filler concentration in the given volume.

The results obtained suggest that the dispersion of the electroconductive particle spacing for a constant concentration of the electroconductive filler is determined by the following factors:
– sizes of particles of the agglomerated electroconductive filler,
– packing factor of the filler particles determined by the physical and chemical structure of the rubber,
– intensity of detachment of single particles from the agglomerate (the deagglomeration process) that depends not only on technology, but also on the agglomerate properties. This parameter affects values of \(\bar{h}\) and \(D\).

It was established that to increase the number of detached particles (to increase the number of contacts and hence to decrease the electric field strength per individual contact), it is better to change the packing factor rather than the detachment intensity. This regularity is connected with the special features of polymer filling [9, 10]. Regulation of the detachment intensity minimizes the dispersion of particle spacing and increases the number of contacts between the conductive particles only if the particles can be distributed over the matrix volume. The increase of the packing factor of the filler particles because of the presence of free high-lying crystalline bands inaccessible for the filler decreases considerably the contribution of the detachment process.
A dependence of the electric durability on the average agglomerate size was studied based on the calculation model presented in [11]. It was established that the increase in the agglomerate size reduces the breakdown voltage for an individual contact (Fig. 1).

The increase of the maximum electric field strength revealed for calculation models in strong electric fields with reduction of the packing factor and sizes of electroconductive filler agglomerate was checked experimentally. The experiments were performed with an aperiodic pulse applied to materials that differed by the packing densities of particles of the filler and its capability to deagglomeration.

We used technical carbon black with different agglomerate sizes, dispersions, and structures having P-514 (ash, intermediately active, with the intermediate dispersion index and intermediate structure index), P-234 (ash, active, with a high dispersion index and an average structure index), and P-366E grades (ash, electroconductive, with a high dispersion and structure indices). The particle packing factor was regulated by the matrix material. Butil Rubber (IIR according to the Standard of the American Society for Testing and Materials (ASTM)) and Styrene Butadiene Rubber (SBR according to ASTM Standard) were used.

Experiments were performed with samples 0.03 m in diameter and 0.05 m in height. The technical carbon concentration was 80 parts by weight (to 100 parts by weight of the rubber). The data presented in Fig. 2 demonstrate that the calculated dependence is confirmed experimentally. Therefore, to operate in strong fields, it is expedient at first to select a polymer based on the effect of the filler particles on packing and then a filler based on the dispersion and structure of the material.

It seems impossible to determine experimentally the packing factor, the particle separation, and other parameters on the microstructure level. Therefore, we adjusted the parameter that can be calculated from electron micrographs of the macrostructure.

To study interrelations of the withstand breakdown voltage with the processes in the electroconductive filler grid, the structure of multicomponent materials was investigated based on the available electron micrographs. Based on the results of research, the structural parameter determined from the texture of micrographs of a multicomponent material was suggested.

FIGURE 1. Dependence of the maximum withstand voltage on the average size of filler particles
(the matrix viscosity was equal to 51 and $\tau = 0.01$ s)

FIGURE 2. Dependence of the maximum withstand voltage on the pulse duration.
Here curve 1 is for IIR, P-366E; curve 2 is for IIR, P-234; and curve 3 is for IIR, P-514
The texture was estimated for a squared area with sizes \((2W + 1) \times (2W + 1)\) with the center at the given point. The data processing algorithm involved the following stages:

1. Formation of flat topological models \(E(j, k)\) taking value 1 for cells with a filler and 0 otherwise.
2. Calculation of arrays of the texture parameter \(T(j, k)\) from the number of brightness gradients in the examined vicinity for each point of the micrograph:

\[
T(j, k) = \frac{1}{(2W + 1)^2} \sum_{m=j-W}^{j+W} \sum_{n=k-W}^{k+W} E(m, n),
\]

where \((j, k)\) are the coordinates of the current point of the micrograph.
3. Construction of histograms of its frequency distribution to calculate \(P_k\).
4. An analysis and quantitative description of the histograms obtained based on the entropy of the texture parameter:

\[
\Delta S_{\text{texture}} = -\sum_{k=0}^{N-1} P_k \log_2 P_k = \Delta S_1.
\]

A relationship of this parameter with the limiting electric durability of materials in strong electric fields (Fig. 2) was established. The entropy of the texture parameter was directly proportional to the maximum withstand voltage of the resistor sample (curve 1: \(\Delta S_1 = 2.9\); curve 2: \(\Delta S_1 = 2.2\); and curve 3: \(\Delta S_1 = 1.8\) with the confidence interval not exceeding 3%).

**CONCLUSION**

Thus, the structure parameter—the entropy of the texture parameter that can be used to estimate the durability of composite materials with polymer components in strong electric fields—has been suggested and substantiated. The results obtained allowed us to propose the two-level method of selection of the material capable of operation in strong electric fields.

**REFERENCES**