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Bifurcation of distribution function of electric breakdown voltages of polymer enamel-lacquer coatings

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Abstract. On the basis of general physics representations, the statistical model of multilayer enamel-lacquer polymer coatings is proposed. Such coatings contain randomly distributed air inclusions. The solution of statistical problem connected with the distribution of electric breakdown voltages of such coatings permits to explain the observed the violation unimodality of histograms of their dielectric strength.

1. Introduction

Investigation of the electric breakdown including its theoretical analysis has a great history. In connection with this, see, for example, the monographs [1–6] where results of investigations on each historical period. From our opinion, despite the absence of new principal approaches to the study of this phenomenon, up to now, it is devoted the great attention for its investigation (see, for example, [7–8]). It is dictated by aspiration to theoretical understanding of this physical phenomenon and by the necessity of practical struggle with its consequences. At this, naturally, the most attention is devoted to the study of electric breakdown in solids which accompanied by degradation of their physical properties including their mechanical destruction.

Currently, we may analyze theoretically the electric breakdown of dielectric and high-resistance semiconductor materials only by phenomenological representations, since consistent realization of such an analysis at microscopic level leads to extremely complicated constructions in frames of statistical physics. It is stipulated not only that the breakdown represents very fast kinetic phenomenon in solid structure which consists of strong connected atoms. And for its description by traditional methods of physical kinetics are unsuitable. It is connected also with the breakdown is a consequence of very strong influence of external electric field on the material such that it cannot be analyzed theoretically by perturbation theory methods. Moreover, the availability of different defects randomly distributed in solid material under consideration influences by correspondent way on kinetics of the electric breakdown. Statistical account of this influence leads to supplement difficulties already at the step of the building of adequate mathematical model, especially in the case when the defect density cannot consider as some kind small value.

Thus, it is natural that theoretical analysis of electric breakdown in solid materials is based, as a rule, on rather rough models of general physics. Each of them is oriented for the description of one significant side of the phenomenon. Our work is devoted to theoretical analysis of the famous experimentally observable effect [9]. It represents that the statistical distribution of random breakdown voltages of polymer enamel-lacquer coatings is not unimodal. The proposed analysis is based on the simple physical model that differs it from the work [10] where an attempt of explanation of this effect is done using only some kind general property of small random samples. In frames of such an approach they cannot to detect the physical reason generated the effect. From our opinion it is not sufficient, since in theoretical physics, it is considered to be that the appearance of nonunimodality should be connected with the presence of the definite physical mechanism generated it. On the basis of our proposed analysis, one may assert that the nonunimodality effect is the consequence of the specific space distribution of defects in the coating film.

2. The phenomenology of electrical breakdown of enamel-lacquer coating

The electrical breakdown is stipulated by ionization with ties breaking between particles of dielectric material directly under action of external electrical field.

The dielectric strength E_{br} of solid dielectrics relative to the electrical breakdown is the relation of breakdown voltage value of dielectric material to the layer thickness of the material in the direction of applied voltage. It lies within relatively narrow limits, namely, $100 \div 1000$ MV/m that is near to E_{br} of strongly compressed gases and very clean liquids.

Presence of defects located by random way in the material and having some random sizes, stipulates, in general case, the random character of breakdown voltages $\tilde{E}$. Statistical character of breakdown voltages is not essential in the case when sample geometric sizes essentially exceed geometric sizes of defects. It leads to smallness of statistical distribution dispersion.

Otherwise, in the case when the pointed out relation between sizes is not such great and the distribution dispersion differs in a noticeable way from zero, one may observe explicitly the statistical spread of breakdown voltages.

At the observation of majority of physical phenomena, such a situation is most common when histograms of the measured physical values, having a statistical spread, possess the unique maximal channel. In mathematical statistics this property of histograms is named *the unimodality*. In connection with this they say about the unimodality of probability distributions describing these random variables. The absence of the unimodality property of statistical distribution points out the presence of some kind special physical mechanism due to which the nonunimodality is broken. Just such a violation of unimodality of breakdown voltages observes when the measurement of the dielectric strength of enamel-lacquer coatings is done.

Let us consider a polymer enamel-lacquer coating without concretization of its chemical composition. It has the form of multilayered film. The preparation technology of such coatings represents of the consistent application of some layers $3 \div 10$ of enamel-lacquer mixture dissolved in a liquid organic solvent. The application of each layer accompanies the appearance of defects in the distribution of polymer substance which have random geometric sizes. These defects change the voltage of electrical breakdown of the layer at the place of their appearance and, correspondingly, they change the breakdown voltage of total multilayered coating. Such defects appear randomly when new lacquer layer is applied. They represent air inclusions. Defects appear due to not predictable adhesion of small air bubbles to the surface of previous dried layer of the film during the application of the subsequent layer of the lacquer. It is essential that at the described physical scenario of defects appearance there is only one air inclusion appearing along the thickness of each new layer.

Thus, defects appearance is a result of pointed out technological process of the formation of film layers. Defects have the form which is similar the semisphere that is stipulated by surface tension of the bubble, if we do not account the wettability effect of air bubble with dry coating surface. At such a case each defect characterizes by one positive random variable, i.e. the bubble radius.

Availability of defects of described type in polymer coating reduces its electrical breakdown voltage since the air dielectric strength is significantly less than dielectric strength of the coating material. Due to randomness of defect radiuses, the electrical breakdown voltage should be also the random variable. Therefore, results of experimental study of dielectric strength of described polymer coatings realized according to definite rule are fixed in the form of statistical histograms of breakdown voltages. On these histograms two maximums are observed [11].

It should be noted that, besides of described mechanism of appearing of random spread of measured electrical breakdown voltages, the randomness may be also as the consequence of randomness of the defect centers distribution in each coating layer. Such a mechanism of randomness may be appeared experimentally in the case of small defect density in each layer and finiteness of the sizes of terminals by which the electrical voltage is applied to coating surface.

Namely, if we denote by l_0 the average distance between defects in the fixed layer and l is the terminal size, then such a situation may be if $l \approx l_0$. Possibility of terminal size influence on the appearance of the nonunimodal distribution has been studied in [12] where it were investigated probability distributions of maximums of small samples of independent positive random variables. It is shown that, despite on small sample volume of random variables that is in the case when the relation l/l_0 is not large, the distribution of its maximum preserve the unimodality of the distribution of corresponding random variables.

In present work we build the statistical model which permits to calculate the probability distribution of breakdown voltage and, in its frames, we give explanation of unimodality of experimentally observed histograms.

3. Statistical mathematical model

Let there be the film of enamel-lacquer coating consisting of N layers of equal thickness d. We number the film layers by $m = 1, ..., N$ in the order of their location in cross section of the film, for example, bottom-up. The base of our statistical model are the point random fields $\{\tilde{x}^{(m)}_{k_m}\}$. Here, the number m is the order number of the coating layer. Points $x^{(m)}_{k_m}$ of random fields correspond to the location of semispheres centers on low surface of each layer $m = 1 \div N$. If the point has the mark $m = 2, ..., N$ then it is located in the plane dividing layers with numbers $m-1$ and m. The points $\tilde{x}^{(1)}_{k_1}$ are located in the plane between the base and the coating. The index k_m, $m = 1 \div N$ in each set $\{\tilde{x}^{(m)}_{k_m}\}$ enumerates points where the defects centers are located in the layer with the number m. With each random point $\tilde{x}^{(m)}_{k_m}$ the positive random variable $r^{(m)}_{k_m} > 0$ is connected, $m = 1 \div N$. Its values are random radiuses of the defect which is located at the point $\tilde{x}^{(m)}_{k_m}$.

Since the density of the defect location is supposed small, we consider that they do not influence to each other in each layer of the coating and, consequently, all air inclusions are statistically independent. Moreover, since each layer is prepared independently from others, it is natural to consider that the defects in different layers are also statistically independent to each other. Thus, we have N copies of statistically equivalent two-dimensional point random fields being statistically independent which are numbered by the index $m = 1 \div N$. Due to the statistical independence of points $\{\tilde{x}^{(m)}_{k_m}\}$ in each random realization with fixed value m, we may consider each such a field as the two-dimensional uniform poissonian one with some kind density $\sigma \sim l_0^{-2}$ of point distribution on the plane. Since all defects are statistically independent, we consider that all random variables $r^{(m)}_{k_m}$, $m = 1 \div N$, $k = 1, 2, 3, ...$ are independent and all of them are equivalent to each other. We denote by $w(r)$ their common probability density.

Let U be the dielectric strength of polymer material and U_0 be the dielectric strength of air.

We represent the trajectory of electronic avalanche by which the electric breakdown is realized in the form of line consisting of subsequently connected segments. Each rectilinear segment connects two centers of defects located in neighboring layers of coating. Let the electronic avalanche moves along the segment which connects the defect center in mth layer with the defect center in $(m+1)$th layer (see, figure 1). Then, on the basis of general physics reasonings, the electrical breakdown voltage of the layer is the sum of electrical breakdown voltage of mth air bubble and electrical breakdown voltage of polymer material along the segment. The first summand is equal $U_0\tilde{r}_m$ according to the definition of the dielectric strength, and the second one is equal $U\cdot(|\tilde{x}^{(m)}_{k_m}-\tilde{x}^{(m+1)}_{k_{m+1}}|-\tilde{r}_m)$. In a result, we have the sum $U_0\tilde{r}_m+U\cdot(|\tilde{x}^{(m)}_{k_m}-\tilde{x}^{(m+1)}_{k_{m+1}}|-\tilde{r}_m)$.

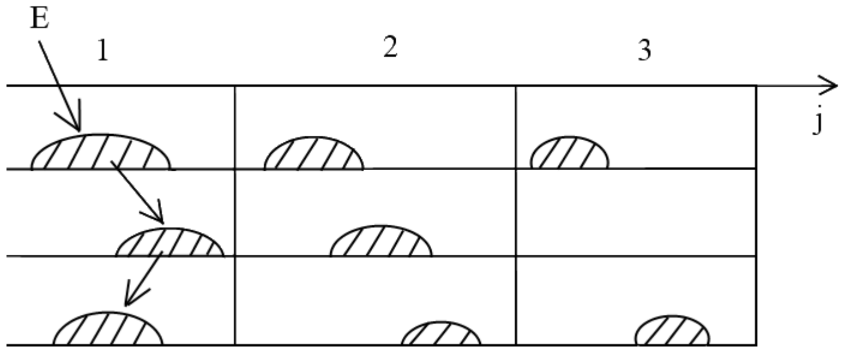

Figure 1. It is shown schematically the three layered coating with defects embedded in them. The movement the electronic avalanche along the trajectory is shown by arrows.

Then the electrical breakdown voltage of the coating is equal

$$\sum_{m=1}^{N}\Big[U_0\tilde{r}_m+U\cdot(|\tilde{x}^{(m)}_{k_m}-\tilde{x}^{(m+1)}_{k_{m+1}}|-\tilde{r}_m)\Big] \tag{1}$$

in the case when the avalanche breakdown is developed along a random trajectory. Here k_1, ..., k_N are the marks of points in random fields with the numbers $m=1,...,N$, $\tilde{x}^{(1)}_{k_1}\equiv\tilde{x}_1$ and $\tilde{x}^{(N+1)}_{k_{N+1}}\equiv\tilde{x}_{N+1}$ is the point on external plane of Nth layer of the coating.

However, it is clear that the avalanche is developed along those trajectory where the sum (1) is minimal. At this, the initial point $\tilde{x}_1$ and the ended one $\tilde{x}_{N+1}$ should be coved by terminals of electrical circuit with voltage applied to the polymer film. For simplicity, we consider that these terminals have square form with the edge length ρ. Centers of these squares are located against each other on opposite planes of the film such that squares are combined at the parallel translation perpendicular to planes of coating.

We denote these squares by Σ for first layer and by Σ' for last one. Thus, the random voltage $\tilde{E}$ of electrical breakdown connected with the random location of defects and their random sizes is given according to (1) by the formula

$$\tilde{E}=\min_{\tilde{x}_1\in\Sigma,\tilde{x}_{N+1}\in\Sigma'}\ \min_{\tilde{x}^{(2)}_{k_2},...,\tilde{x}^{(N)}_{k_N}}\sum_{m=1}^{N}\Big[U_0\tilde{r}^{(m)}_{k_m}+U\cdot(|\tilde{x}^{(m)}_{k_m}-\tilde{x}^{(m+1)}_{k_{m+1}}|-\tilde{r}_m)\Big]. \tag{2}$$

The problem consists of the determination of the probability distribution density $f(E)$ of this random variable if the density σ characterizing random fields $\{\tilde{x}^{(m)}_{k_m}\}$, $m=1\div N$, and the probability distribution density $w(r)$ are fixed. At this, we consider that the average size $\langle \tilde{r} \rangle = \int_0^\infty r w(r) dr$ of the defect is much smaller than l_0 such that defects in each layer do not overlap with a suppressing probability.

It is clear that the statistical problem pointed out, despite of its formulation transparency, is extremely complicated for the analysis, in view the necessity of enumeration of the continuum of possible trajectories along which the electrical breakdown may be developed with the account of their possible contribution in desired probability. Due to this reason, we construct now more simple mathematical model on the basis of discretization of the described one. It allows to do its relatively simple mathematical analysis.

We divide external plane of the film by $l \times l$ square net such that its lines are parallel to corresponding sides of squares Σ and Σ'. Thus, we assume that the edge length of each square is equal to l which satisfies the conditions $\langle \tilde{r} \rangle \ll \rho \ll l_0$. We introduce the coordinate description of the net squares by pairs of integers $\langle i, j \rangle \in \mathbf{Z}^2$. After that, we build planes through lines generating square net on external film surface such that they are perpendicular to the film. These planes divide the film by parallelepipeds. Squared bases of parallelepipeds are located in opposite flat surfaces of the film. We name these parallelepipeds the *channels*. They are cut by parallel planes which are borders of the film layers. In a result, each channel are divided on same parallelepipeds with squared $l \times l$ bases located on planes which limit the film layers and their heights are equal d. Each such a parallelepiped we name the *cell*. Thus, each channel is split into N same cells put on each other. Each cell in the film is identified by triple of numbers, namely, the pair $\langle i, j \rangle$ and the number $m = 1 \div N$ of the film layer where this cell is located.

Each cell characterizing by the triple $\langle i, j, m \rangle$ may contain no more that one defect and the probability of such an random event is equal $v < 1$. Such a situation is provided by inequalities $\langle \tilde{r} \rangle \ll l \ll l_0$. The value of the probability v is determined by choosing of the ratio l/l_0 and v is the free parameter of the model. The random radius of the semisphere of defect containing in this cell we denote by $\tilde{r}_{i,j}^{(m)}$.

Since random point field $\{\tilde{x}_{k_m}^{(m)}\}$ are poissonian one, i.e. all their points are statistically independent, we consider that all random events of hit or missing defects centers in each cell are statistically independent.

Thus, after the conducted discretization, we have conversed random point fields $\{\tilde{x}_{k_m}^{(m)}\}$ on borders of layers into random point fields on discrete set with points describing by the collections $\langle i, j, m \rangle$.

Since random sizes $\tilde{r}_{k_m}^{(m)}$ in the initial model are not statistically depend on field points $\{\tilde{x}_{k_m}^{(m)}\}$ at each value $m = 1 \div N$, it is natural to suppose that just as well all random variables $\tilde{r}_{i,j}^{(m)}$, $\langle i, j \rangle \in \mathbf{Z}^2$, $m = 1, ..., N$ are statistically independent. These random variables are identically distributed with the density $w(\cdot)$.

Now we consider that the value l is chosen by such a way that the breakdown electronic avalanche is developed in a fixed channel and it cannot penetrate during movement in neighbor channels. Moreover, we consider that $l \ll d$. It permits to neglect by difference between the length of each trajectory segment and the thickness d of the film layer. Therefore, it is allowed to place points $\tilde{x}_{k_1}^{(1)}$ and $\tilde{x}_{N+1}$ in centers of bases of corresponding channels and to consider them not random.

At such assumptions the sum (1) in the channel defined by the pair $\langle i, j \rangle$ is equal

$$U_0 \tilde{r}_{i,j} + U(Nd - \tilde{r}_{i,j}) = UNd - (U - U_0)\tilde{r}_{i,j}$$

where random variables

$$\tilde{r}_{i,j} \;=\; \sum_{m=1}^{N} \tilde{r}_{i,j}^{(m)}$$

represent summed "sizes" of air inclusions in each channel defined by the pair $\langle i, j \rangle$ and consisting of N subsequently laid cells. Then the formula (2) is written in the form

$$\tilde{E} = \min_{\langle i,j \rangle \in \Sigma} [UNd - (U - U_0)\tilde{r}_{i,j}] = UNd - (U - U_0) \max_{\langle i,j \rangle \in \Sigma} \tilde{r}_{i,j}$$

where the minimization on $\tilde{x}_1$ is replaced by the minimization on all channels which coordinates are located in the domain coved by terminal of applied electrical voltage, and the minimization on $\tilde{x}_{N+1}$ is not necessary, since the upper cell of each channel is defined by the choosing of their lower cell. The necessity of the internal minimization that is present in the formula (2) is not needed since corresponding trajectories transform in straight segments with initial points $\langle i, j, 1 \rangle$.

Thus, in the simplified model, the electrical breakdown voltage is determined completely by difference of electrical breakdown voltage of the polymer material without defects and electrical breakdown voltage of the air layer having the thickness equal to the sum of all sizes of air inclusions.

Since random variables $\tilde{r}_{i,j}^{(m)}$, $\langle i, j \rangle \in \mathbf{Z}^2$, $m = 1 \div N$ are independent, then the random variables $\tilde{r}_{i,j}$ are also independent. Introducing the value $E_0 = UNd$ of electrical breakdown voltage of the polymer material and the positive difference $\nu = U - U_0$ of dielectric strengths of polymer and air, we write down the basic formula for random variable $\tilde{E}$ in the form

$$\tilde{E} = E_0 - \nu \max_{\langle i,j \rangle \in \Sigma} \tilde{r}_{i,j} \tag{3}$$

where random variables $\tilde{r}_{i,j}$ are statistically independent and equivalently distributed. Each of them represents the sum of independent random variables equivalently distributed with the density $w(r)$.

Linear size ρ of electric terminals exceeds in significant way the linear size l. Consequently, the electronic avalanche may be realized in any channels covered by the square Σ. It is realized namely in that channel where the difference between sum of thicknesses of polymer material of all cells in the channel and sum of defect sizes in them is minimal. Therefore, it is necessary to calculate the maximum on all channels when the random variable $\tilde{E}$ defining. Using (3), the probability $\Pr\{\tilde{E} < E\}$ of the electrical breakdown origin in the case when $\tilde{E}$ exceeds some kind voltage E reduces to the determining of the probability distribution of the sample maximum of independent and equivalently distributed variables

$$\Pr\{\tilde{E} < E\} = \Pr\left\{\max_{i,j} \tilde{r}_{i,j} > s\right\}$$

where $s = (E_0 - E)/\nu$, since the inequality $E > E_0 - \nu \max_{i,j} \tilde{r}_{i,j}$ is equivalent $s < \max_{i,j} \tilde{r}_{i,j}$. Then

$$\Pr\left\{\max_{\langle i,j \rangle \in \Sigma} \tilde{r}_{i,j} > s\right\} = 1 - \Pr\left\{\max_{\langle i,j \rangle \in \Sigma} \tilde{r}_{i,j} < s\right\}$$

where we have used the supposition about the continuity of probability distribution of typical random variable $\tilde{r}_{i,j}$ at $s > 0$.

Using properties of statistical independence of random variables $\tilde{r}_{i,j}$ equivalently distributed with random variable $\tilde{r}_{0,0}$, we find

$$\Pr\{\tilde{E} < E\} = 1 - (\Pr\{\tilde{r}_{0,0} < s\})^M$$

where $s \geq 0$, since we consider that $E > E_0$, and also we introduce the designation $M = |\Sigma|$ of the channels number with bases located in Σ.

Consequently, the distribution density of the random variable $\tilde{E}$ is equal

$$f(E) = \frac{\mathrm{d}}{\mathrm{d}E}\Pr\{\tilde{E} < E\} = \nu^{-1}M \cdot [Q_N(s)]^{M-1} \cdot q_{_N}(s)|_{s=(E_0-E)/\nu} \tag{4}$$

where it is taken into account that $\mathrm{d}s/\mathrm{d}E = -\nu^{-1}$, and we introduce the designations of the probability $Q_N(s) = \Pr\{\tilde{r}_{0,0} < s\}$ and its density

$$q_{_N}(s) = \frac{\mathrm{d}}{\mathrm{d}s}Q_N(s)\,. \tag{5}$$

The unimodal distributions of the random variable $\tilde{r}_{0,0}$ which are contained in the definite class of such distributions have been applied for the study of the density $q_{_N}(s)$ in the article [12]. It is shown that the availability of the multiplier $[Q_N(s)]^{M-1}$ in the formula (4), generally speaking, does not lead to violation of unimodality of the distribution density $f(E)$ when $q_{_N}(s)$ is unimodal density of this class. Therefore, one may attempt to explain the unimodality violation of the electrical breakdown voltages distribution of multilayered film by the unimodality violation of the density $q_{_N}(s)$. In present work we solve namely this problem.

First of all, we shall find the explicit formula for this density. We note that due to practical reasons, the number M does not very large. Therefore, make no sense to apply the limit distribution laws of the maximum of the sum of independent equivalently distributed variables, which correspond the case when $M \to \infty$.

The probability $Q_N(s)$ consists of the sum of probabilities of random events $\{\tilde{r}_{0,0} < s$, and k cells contain defects$\}$ which are the product of independent at $k > 0$ events $\{\tilde{r}_{0,0} < s\}$ and $\{k$ cells contain defects$\}$. The probability of last event which is connected with the sequence of independent trials is equal

$$\Pr\{k \text{ layers contain inclusions}\} = \binom{N}{k}(1-v)^{N-k}v^k\,.$$

At $k = 0$ the conditional probability $\Pr\{\tilde{r}_{0,0} < s|k = 0\}$ is equal to the Heviside function $\theta(s)$, since in this case $\tilde{r}_{0,0} = 0$. Then, using the formula of complete probability, we obtain the following expression

$$Q_N(s) = \Pr\{\tilde{r}_{0,0} < s\} = (1-v)^N\theta(s) + \sum_{k=1}^{N}\binom{N}{k}v^k(1-v)^{N-k}\Pr\{\tilde{r}_{0,0} < s|k \neq 0\}\,. \tag{6}$$

Thus, the desired probability is defined completely by the probability distribution of typical random variable $\tilde{r}_{0,0}$. The last represents the probability distribution of the sum of k independent equivalently distributed with the density $w(r)$ random variables. Therefore (see, for example, [13]), it is equal

$$\Pr\{\tilde{r}_{0,0} \leq s|k \neq 0\} = \int_0^s \underbrace{(w * \ldots * w)}_{k}(r)\mathrm{d}r \tag{7}$$

where the symbol $*$ denotes the binary convolution operation of probability distributions. For two arbitrary densities g_1 and g_2 of nonnegative random variables this operation is defined by the formula

$$(g_1 * g_2)(r) = \int_{0+}^{r} g_1(r')g_2(r-r')\mathrm{d}r'\,. \tag{8}$$

In a result, the distribution density $q_N(s)$ is given, according to (5–7), by the following formula

$$q_N(s) = (1-v)^N \delta(s) + \sum_{k=1}^{N} \binom{N}{k} v^k (1-v)^{N-k} \underbrace{(w * ... * w)}_{k}(s) \qquad (9)$$

where we take into account that $\mathrm{d}\theta(s)/\mathrm{d}s = \delta(s)$ is the Dirac function. We note that this formula permits the write in more compact form of Nth degree of the convolution operation

$$q_N(s) = (\delta(r)(1-v) + vw(r))_*^N(s)\,. \qquad (10)$$

Availability of δ-functional singularity at the point $s = 0$ in the distribution density $q_N(s)$ expression leads to the availability of δ-functional singularity at the point $E = E_0$ of the distribution density $f(E)$ that gives the maximum on histograms of experimental data. Thus, for availability of the probability distribution nonunimodality of electrical breakdown voltages, it is sufficient to show that the distribution density $q_N(s)$ has, at least, one more top at some kind point $s \neq 0$.

4. Analysis of the statistical model

In this section we show that, in the case when typical sizes of defects are very small in comparison with d and the values of parameter v are not very small, the density $q_N(s)$ has the top at nonzero point.

In practice, the type of the probability density

$$w(r) \;=\; \frac{\mathrm{d}}{\mathrm{d}r}\Pr\{\tilde{r} < r\}\,, \qquad \int\limits_0^d w(r)\mathrm{d}r \;=\; 1$$

and its parameters as well as the probability v are not known. Therefore, at the theoretical analysis, it is necessary to operate with some kind model density and to choose the values of model parameters by means of processing of statistical experimental data. Then, it is reasonable to analyze possibilities of qualitative behavior of the distribution density $f(E)$ using some kind rather wide class of probability distributions with the aim of finding of adequate density in frames of our general model.

Following the aim of present work, it is necessary to find such type of model densities $w(r)$ which lead to the violation of unimodality of the distribution density $f(E)$ or, that is the same, to the violation of unimodality of the distribution density (10).

In the model, proposed in the work [10], it was supposed that $w(r) = w_0 e^{-\lambda r}$ at $0 < r < d$, $\lambda \sim \langle \tilde{r} \rangle > 0$ and out of the segment $[0, d]$ the density $w(r)$ is equal to zero, where $w_0 = \lambda(1 - e^{-\lambda d})^{-1}$. In this case, one may represent the formula of density in the form

$$w(r) = w_0 u(r) e^{-\lambda r}\,, \quad u(r) = \theta(d-r)\theta(r) \qquad (11)$$

where $\theta(\cdot)$ is the Heviside function defined as $\theta(r) = \{1 \text{ at } r \geq 0; 0 \text{ at } r < 0\}$.

We shall analyze the case when λd is very large value such that $\langle \tilde{r} \rangle = \lambda^{-1} \ll d$. Then, we may consider that $w(r)$ is exponential. The using of such a distribution density is justified by the fact that defects with very small sizes are most probable, namely, the probability $P(r)$ of the defect generation which has the size less than r should be $P(r) = 1 - \lambda r + o(r)$ at $r \to 0$. Therefore, further we suppose that $w(r) = \lambda \exp(-\lambda r)$. For the density of such a type, on the basis of the definition (8), we have

$$\underbrace{(w * ... * w)}_{k}(s) = \lambda^k e^{-\lambda s} \int_0^s \mathrm{d}r_1 \int_0^{r_1} \mathrm{d}r_2 ... \int_0^{r_{k-2}} \mathrm{d}r_{k-1} = \frac{\lambda^k s^{k-1}}{(k-1)!} e^{-\lambda s}\,.$$

Thus, according to (9), at $s > 0$ we have

$$q_N(s) = \exp(-\lambda s) R_N(s)\,, \qquad R_N(s) = \sum_{k=1}^{N} \frac{s^{k-1}}{(k-1)!} \binom{N}{k} (v\lambda)^k (1-v)^{N-k}\,. \tag{12}$$

Our analysis of unimodality of the density (11) is based on the following simple algebraic statement.

Theorem 1. *Let $P_N(s)$ be the polynomial of N degree which has the form $P_N(s) = s^n P_{1,N-n}(s) - P_{2,n}(s)$ where polynomials $P_{1,N-n}(s) = a_0^{(1)} s^{N-n} + ... + a_{N-n}^{(1)}$ and $P_{2,n}(s) = a_0^{(2)} s^n + ... + a_n^{(2)}$ have degrees $N-n$ and n, correspondingly, $a_0^{(1)} > 0$, $a_0^{(2)} > 0$ and all coefficients of them are nonnegative. Then the polynomial $P_N(s)$ has the unique root at $s > 0$.*

Proof. Let $P_N^{(j)}(s) = [s^n P_{1,N-n}]^{(j)} - P_{2,n}^{(j)}(s)$, $j = 1 \div n$ be the derivatives of the polynomial $P_N(s)$ up to the nth degree inclusively. According to the definition of this polynomial, $P_N^{(j)}(0) \le 0$, $j = 1 \div n$. Besides, $P_N^{(n)}(0) = -a_0^{(2)} n! < 0$. Consequently, each derivative decreases in the neighborhood of the point $s = 0$ at $s > 0$. Besides, $P_N^{(j)}(s) \to \infty$ at $s \to \infty$ with $j = 0, 1, ..., n$, since the asymptotical behavior of these derivatives has the following form $P_N^{(j)}(s) \sim [s^n P_{1,N-n}(s)]^{(j)} \sim a_0^{(1)} [N!/(N-n)!] s^{N-n}$. Therefore, each of these derivatives has a root at $s > 0$.

Let us prove using the consistent descent from of the largest order $j = n$ of derivative to the order $j = 1$, that this root is unique for each function $P_N^{(j)}(s)$.

At $j = n$, due to positivity of coefficients of the polynomial $P_{1,N-n}(s)$, the polynomial $P_N^{(n)}(s)$ increases at $s > 0$. Therefore, it has the unique root. But in this case, the polynomial $P_N^{(n-1)}(s)$ has the unique minimum at $s > 0$, which is less than zero, since it decreases in the neighborhood of the point $s = 0$. Then, it also has a unique root at $s > 0$.

Suppose, we have proved the uniqueness of the root of the derivative $P_N^{(j)}(s)$. Then the function $P_N^{(j-1)}(s)$ has the unique minimum at $s > 0$ and it decreases at sufficiently small positive values s. In this case, the minimum is negative and, therefore, this function has a unique root at $s > 0$. Since j is arbitrary, we have proved that each derivative of order $j = 0, 1, ..., n$ has a unique root at $s > 0$. In particular, the polynomial $P_N(s)$ has such a property. *End of proof.*

Now, using (12), we differentiate the density $q_N(s)$:

$$q'_N(s) = \exp(-\lambda s)[R'_N(s) - \lambda R_N(s)]\,, \qquad R'_N(s) = \sum_{k=2}^{N} \frac{s^{k-2}}{(k-2)!} \binom{N}{k} (v\lambda)^k (1-v)^{N-k}$$

where

$$R'_N(s) - \lambda R_N(s) = -\frac{\lambda}{N!} (v\lambda)^N s^{N-1} +$$
$$+\lambda \sum_{k=0}^{N-2} \frac{s^k}{k!} \frac{(v\lambda)^{k+1}(1-v)^{N-k-2}}{(k+2)} \binom{N}{k+1} [(N-k-1)v - (1-v)(k+2)]\,. \tag{13}$$

At $s > 0$ the equation $q'_N(s) = 0$ for extremal points of the density $q_N(s)$ is reduced, on the bases of (13), to the equation $\lambda R_N(s) - R'_N(s) = 0$. Then, if the inequality

$$(N-k-1)v - (1-v)(k+2) < 0, \tag{14}$$

takes place for all $k = 0, 1, ..., N-2$, coefficients at all degrees s^k, $k = 0, 1, ..., N-2$ are negative. Therefore, the density $q_N(s)$ decreases at $s > 0$. Consequently, it is unimodal with the top in $s = 0$.

If parameters v and N such that it is realized the opposite inequality at some values $k = 0 \div N-2$, then it is possible the appearance another nonzero top. It is important to note that the parameter λ is not present in the inequality (14) and, consequently, the unimodality violation does not depend on defect sizes in frames of the model under consideration.

From (14) it follows that $(N+1)v < k+2$. Consequently, it takes place for all k if $v < 2/(N+1)$. Besides, if $v > N/(N+1)$, then $k+2 > N$, that is impossible, and therefore, the opposite inequality $(N-k-1)v-(1-v)(k+2) > 0$ takes place at these values v and at all permissible values k. Thus, there is such a value k_* that coefficients of the polynomial $[R_N(s) - \lambda R'(s)]$ are positive when $k \leq k_*$, besides the coefficient at the term s^{N-1}, and they are negative at $k > k_*$. Applying the above algebraic statement, one may assert that the density $q_N(s)$ is unimodal at $v < 2/(N+1)$, and it has another one supplementary top at $v > 2/(N+1)$.

5. Conclusion

On the basis of general physics presentations, the statistical model has been built such that it permits to find the physical reason of appearance of more than one top in the distribution of electrical breakdown voltages. At this, the resulting breakdown voltage represented by the minimum of independent equivalently distributed random breakdown voltages of small regions of the polymer film with such typical sizes that they are comparable with average distance l_0 between air inclusions in layers of coating. In turn, the average distance l_0 much more exceeds both average defect size and the characteristic width l of electronic avalanche that realizes the electrical breakdown.

It is shown that, at condition of smallness of average defect size in comparison with thickness d of layer, the violation of unimodality of electrical breakdown voltages distribution of dielectric enamel-lacquer coatings is connected with that it may be located no more than one defect in the layer of coating and the distribution density σ of defects is sufficiently small. At this, one of tops of the density $f(E)$ coincides with the breakdown voltage E_0 of the material without defects.

6. References

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