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DISTRIBUTION OF THE ELECTRIC FIELD IN A POLYMER FILM UNDER A SHORT-TERM ACTION OF A HIGH-VOLTAGE PULSE

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Abstract. Calculations of the electric field distribution in the electrode system in the form of a spherical concentric capacitor simulating a micro-tip on the cathode were performed under the action of a high-voltage pulse with a front steepness of about 1 GV/s and electron injection from the micro-tip. The penetration depth of the negative space charge (NSC) into the polymer was shown to be 0.2–0.3 μm during a 150–250 ns pulse front edge. Electrical overvoltages caused by the geometry of the electrode system and the accumulating NSC occurred in the NSC accumulation region at the pulse front edge. The field strength at the cathode decreased multiple times during the transition from the pulse front to its plateau in 100–200 ns.

Keywords: high voltage pulse, space charge, field strength, redistribution of the electric field

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РАСПРЕДЕЛЕНИЕ ЭЛЕКТРИЧЕСКОГО ПОЛЯ В ПОЛИМЕРНОЙ ПЛЕНКЕ ПРИ КРАТКОВРЕМЕННОМ ВОЗДЕЙСТВИИ ВЫСОКОВОЛЬТНОГО ИМПУЛЬСА

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Аннотация. Выполнены расчеты распределения электрического поля в электродной системе в виде сферического концентрического конденсатора, моделирующего микроострие на катоде, при воздействии высоковольтного импульса с крутизной фронта около 1 ГВ/с и инжекции электронов с микроострия. Показано, что при длительности переднего фронта импульса 150 – 250 нс глубина прорастания отрицательного объемного заряда (NSC) в полимер составляет 0,2 – 0,3 мкм. Электрические перенапряжения, обусловленные геометрией электродной системы и накапливающимся NSC, возникают в области накопления NSC на переднем фронте импульса. Напряженность поля у катода падает кратно при переходе с фронта на плато импульса за 100 – 200 нс.

Ключевые слова: высоковольтный импульс, объемный заряд, напряженность поля, перераспределение электрического поля

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Introduction

In recent years, considerable attention has been paid to the electrical strength of polymer dielectrics exposed to pulse voltages [1–6]. A growing number of studies address various aspects of the impact of pulsed voltages on polymer materials, specifically, accumulation of space charge [6–8], influence of temperature [9] and rise rate [10, 11]. Various physical mechanisms are also proposed to explain the characteristics of pulse breakdown in polymers [12–15]. However, in our opinion, a physically meaningful interpretation of the experimental results and proposed models is impossible without estimating the magnitude of the realistically achievable electric field strength, its distribution in the polymer, and variation over time under the pulse voltage conditions.

Such estimations should be carried out bearing in mind that polymer dielectrics are characterized by a high concentration of trap states that form a space charge. Charges injected from the electrodes are captured in these traps. Accumulation of the space charge, in turn, significantly affects the distribution of the electric field, limiting the injection current and producing local areas of electrical overvoltage.

We should note that electrons and holes in a strong electric field are injected into the polymer from micro-protrusions on the electrodes; the electric field strength F at the tips of these protrusions exceeds the average value $F_{av} = U/d$ (U is the voltage across a sample with a thickness d).

Unfortunately, modern acoustic methods for probing space charge and electric fields in polymers cannot be used to measure the localization of the charge over the sample surface and do not provide the required depth resolution, which should be at least 0.1 μm for micron-thick polymer films [16]. Consequently, numerical methods are widely used to solve this problem, but calculations are generally limited to steady-state distributions of fields and charges [17, 18].

The goal of this study is to calculate the distributions of the electric field near a micro-tip in a polymer dielectric accumulating negative space charge (NSC), exposed to steep-front electric pulses. An additional goal was to estimate the magnitude of local overvoltages occurring at the injecting electrodes and at the boundary where NSC penetrates into the polymer.

Problem statement

The micro-tip on the surface of the cathode was modeled by an electrode system consisting of a spherical concentric capacitor, where an electrode of small radius r_c was regarded as a micro-tip, and a sphere of larger radius R as an anode. The diagram of the electrode system is shown in Fig. 1. Such systems are widely used to model micro-tips with constant surface curvature [19].

The advantage of this model is the simplicity of the mathematical equations used to describe the distribution of the electric field.

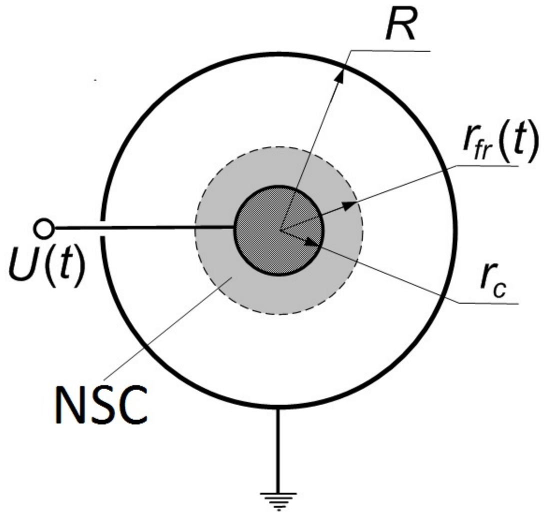

Fig. 1. Diagram of electrode system configured as spherical concentric capacitor: R, r_c are the radii of the outer and inner spheres, respectively; $r_{fr}(t)$ is the NSC penetration depth into the polymer at time t

A large drawback, however, is that the model is not fully consistent with the real conditions, satisfactorily reproducing them only at distances at which the electric field strength F exceeds its average value F_{av} (F_{av} is understood here as the field in a planar sample with a thickness $d = R - r_c$).

The distribution of the electric field strength $F(r,t)$ in the electrode gap of the system at any given time t is described by the Poisson equation, taking the following form in spherical coordinates:

$$\frac{d}{dr}F(r,t)+\frac{2}{r}F(r,t)=\frac{\rho(r,t)}{\varepsilon\varepsilon_0}, \qquad (1)$$

where ε is the relative permittivity of the polymer dielectric; ε_0 is the electric constant, $\varepsilon_0 = 10\cdot 8.85^{-12}$ F/m; e is the electron charge, $e = -1.6022\cdot 10^{-19}$ C; $\rho(r,t)$ is the instantaneous NSC density, determined by the concentrations of free and trapped electrons.

Since electrical breakdown mainly occurs at the leading edge of the pulse and much less frequently at the plateau or at the trailing edge [12, 14], we primarily focused on calculations of the field distribution over the time interval covering the leading edge and the initial plateau of the high-voltage pulse. In view of this, we assumed in the calculations that the voltage U across the sample varies as follows over time:

$$U(t)=\begin{Bmatrix}\frac{dU}{dt}t, \text{ for } t\le\Delta \\ U_{amp}, \text{ for } t>\Delta\end{Bmatrix}. \qquad (2)$$

Here t is the time; U_{amp} and Δ are the amplitude and rise time of the pulse, respectively.

The voltage rise rate across the sample (i.e., the steepness of the leading edge) is $dU/dt = U_{amp}/\Delta$.

In accordance with the recommendations given in [20], we assume that injection of electrons from the cathode into the polymer dielectric in a strong electric field is induced by field electron emission, while the dependence of the current density of this emission on the field strength at the cathode is determined by the Fowler—Nordheim equation.

If the potential barrier at the cathode/dielectric interface is assumed to be triangular, then the equation for calculating the injection current density $j_c(t)$ can be written as

$$j_c=\frac{e^3F_c^2(t)}{16\pi^2\hbar\Delta_e}\exp\left(-\frac{4\sqrt{2m}\Delta_e^{3/2}}{3\hbar F_c(t)e}\right), \qquad (3)$$

where m, kg, is the electron mass; $\hbar$, J·s, is the Planck constant; $F_c(t)$, V/m, is the time-variable field strength at the cathode; Δ_e, eV, is the potential barrier height at the cathode/polymer interface.

Note here that Eq. (3) is simplified and does not take into account the decrease in potential barrier height in a strong electric field.

The value of Δ_e for injection of electrons into the polymer from a metal cathode can be defined as the difference between the cathode work function and the electron affinity A_c of the polymer dielectric. The work function for metals is 2.0—5.5 eV, while A_c for polymer dielectrics typically lies in the range of 1—2 eV, so the potential barrier height Δ_e can be estimated as 1—4 eV. This result is consistent with the known experimental data. For example, the potential barrier heights at the interfaces of polyethylene terephthalate (PET) with aluminum and copper were experimentally determined in [21] to be 2.8 and 2.9 eV, respectively.

The solution of Eq. (1) takes the form

$$F(r,t)=\left(\frac{r_c}{r}\right)^2\left[F_c(t)+\frac{1}{\varepsilon\varepsilon_0 r_c^2}\int_{r_c}^{r}x^2\rho(x,t)dx\right]. \tag{4}$$

Bearing in mind that

$$\int_{r_c}^{R}F(r,t)dr=U(t), \tag{5}$$

and substituting expression (4) into relation (5), we obtain that the dependence of the field strength at the cathode on time follows the expression

$$F_c(t)=\frac{RU(t)}{r_c(R-r_c)}-\frac{R}{\varepsilon\varepsilon_0 r_c(R-r_c)}\int_{r_c}^{R}\left[\int_{r_c}^{r}x^2\rho(x,t)dx\right]\frac{dr}{r^2}. \tag{6}$$

Let us denote the total charge enclosed between two spherical surfaces with radii r_c and r at time t as

$$q(r,t)=4\pi\int_{r_c}^{r}x^2\rho(x,t)dx, \tag{7}$$

In view of notation (7) and substituting expression (6) into solution (4), we obtain:

$$F(r,t)=\frac{r_c RU(t)}{r^2(R-r_c)}-\frac{r_c R}{4\pi\varepsilon\varepsilon_0 r^2(R-r_c)}\int_{r_c}^{R}\frac{q(r,t)}{r^2}dr+\frac{q(r,t)}{4\pi\varepsilon\varepsilon_0 r^2}. \tag{8}$$

Dependence (8) allows to calculate the distribution of the electric field in the electrode gap at any given time. We should note here that the ratio $U(t)/(R-r_c)$ can be taken as a formula for field strength in a planar sample of flat geometry with a thickness $d=R-r_c$.

It follows from Eq. (8) that the electric field strength at any distance r from the cathode is a superposition of three fields:

the field determined by the geometry of the electrode system (the first term);

the field generated by the space charge distributed throughout the sample (the second term);

the field generated by the NSC distributed in the space between the cathode and the surface of radius r (the third term).

The following equality holds true for the polymer exposed to a short electrical pulse during which the electrons do not have sufficient time to reach the opposite electrode (anode) and interact with it:

$$q[r_{fr}(t)]=Q(t),$$

where $Q(t)$ is the total charge accumulated in the electrode gap by time t due to injection of electrons from the cathode; recall that $r_{fr}(t)$ is the electron penetration depth into the polymer.

The total charge $Q(t)$ follows the expression

$$Q(t)=4\pi r_c^2\int_0^t j_c(t')dt'. \tag{9}$$

The charge penetration depth into the polymer at time t is defined as

$$r_{fr}(t)=\mu_{dr}\int_0^t F_{fr}(t')dt', \tag{10}$$

where $F_{fr}(t)$ is the field strength at the boundary where the charge penetrates into the polymer (depending on time); μ_{dr} is the electron drift mobility in the polymer dielectric.

The quantity μ_{dr} takes on a small value, estimated to be about 10^{-9} m^2/(V·s) even in a strong constant electric field (at $F_{av} \approx 10^8$ V/m) [22].

The exact analytical expression for the function $q(r,t)$ is unknown and is unlikely to be determined, since it requires taking into account the electron trapping and detrapping rates at any r and t.

Evidently, the form of function $q(r,t)$ depends on many factors:

injection current density,

charge carrier mobility,

voltage across electrodes,

energy density of trapped states,

duration of electrical pulse.

However, according to the definition, the function $q(r,t)$ increases monotonically for $r_c \le r \le r_{fr}$ as r increases, while $q(r_c,t) \equiv 0$. For $r > r_{fr}$, $q(r,t)$ does not depend on the coordinate and $q(r,t) = Q(t)$.

When a short pulse is applied to the polymer dielectric, the electrons injected from the cathode are trapped and retained in traps, generating the NSC.

The average electron trapping time is expressed as

$$t_{trap} = \tau_0 \cdot \exp[E_{trap}/(k_B T)],$$

where E_{trap} is the trap depth, T is the temperature, k_B is the Boltzmann constant; τ_0 is the characteristic time, $\tau_0 \approx 0.1$ ps.

For a pulse lasting 10 ns, electrons remain in traps with a depth of more than 0.3 eV for the entire duration of the pulse at room temperature. It was found in [11] that the density of the NSC penetrating into the polymer can be considered constant for an electric pulse duration from 10 to 1000 ns and an exponential trap depth distribution. In this case, the expression for calculating the function $q(r,t)$ takes the form

$$q(r,t) = \frac{r^3 - r_c^3}{r_{fr}^3(t) - r_c^3} Q(t). \tag{11}$$

Relation (11) should be regarded as an approximation that can be used to calculate the field distribution near the micro-tip. However, in practice, the boundary of the injected charge is not so sharply pronounced, and the charge distribution density should still depend on the coordinate. Therefore, the function $q(r,t)$ for $r \le r_{fr}$ is approximated in [23] by a power series with the base $r - r_c$.

To use a similar approximation method for $q(r,t)$, we assume that the function $q(r,t)$ takes the form

$$q(r,t) = a(t)(r - r_c) + b(t)\left(r - r_c\right)^2. \tag{12}$$

The coefficients of the polynomial $a(t)$ and $b(t)$ for time t can be found from the conditions

$$q[r_{fr}(t)] = Q(t) \text{ and } dq(r,t)/dr = 0 \text{ for } r = r_{fr}.$$

This approximation corresponds to the condition that the NSC density is zero at the NSC penetration front, where $a(t)$ and $b(t)$ are defined as

$$\left\{\begin{aligned} b(t) &= \frac{Q(t) - a(t)\left[r_{fr}(t) - r_c\right]}{\left[r_{fr}(t) - r_c\right]^2} \\ a(t) &= -2b(t)\left[r_{fr}(t) - r_c\right] \end{aligned}\right\}^2. \tag{13}$$

Notably, in the simplest case, $q(r,t)$ can be approximated by a straightforward linear function

$$q(r,t)=\frac{r-r_c}{r_{fr}(t)-r_c}Q(t). \tag{14}$$

The dependence determined by relation (13) corresponds to the case $\rho(r,t) \sim r^{-2}$, where $\rho(r_{fr},t) \neq 0$.

Substituting relations (11), (12) and (14) into expression (8) yields analytical expressions for calculating the electric field strength in the electrode gap at a fixed t. The time argument of the function $F(r,t)$ is determined by the time dependence of the quantities $U(t)$, $Q(t)$ and $r_{fr}(t)$, which can be regarded as integral parameters in relation (8), independent of the coordinate. An additional factor complicating the calculation of the dependence $F(r,t)$ is that the charge penetration depth $r_{fr}(t)$ is related to the field strength $F_{fr}(t)$ at the NSC boundary by the integral in expression (10).

Thus, the process of NSC accumulation is a self-consistent problem. For this reason, the dependences $F_c(t)$ and $F_{fr}(t)$ were calculated using a time-stepping method. The method was applied assuming that the field and charge distributions remained constant during the discretization time step Δt (the value $\Delta t = 1$ ns was adopted in the calculations). F_c and F_{fr} were calculated at the ith step at time t_i based on the values of these quantities calculated at the previous time step. The integrals in relations (9) and (10) determining the time variation of the quantities $Q(t)$ and $r_{fr}(t)$ were replaced by sums.

The calculations accounted for the fact that intense field electron emission from the microtip occurs at the cathode when a certain critical value of the electric field strength is reached (in accordance with Eq. (3)). As a result, electrons in a weak electric field are practically not injected into the polymer, NSC does not accumulate, and the distribution of the electric field in the electrode gap is determined by the geometry of the electrode system (see the first term in Eq. (8)). Therefore, the first step ($i = 1$) in the calculation of $F(r,t)$ was not the time $t = 0$, corresponding to the beginning of the voltage increase, but the time t_1, when the voltage across the sample reached the value $U(t_1)$, at which a noticeable injection current occurred.

The following initial conditions were adopted for calculations at the first time step:

$$Q(t_1) = 0,\ r_{fr}(t_1) = r_c \text{ and } F_{fr}(t_1) = F_c(t_1).$$

The value of t_1 was 30–40 ns (depending on the amplitude and rise time of the pulse).

Numerical calculations were performed for the following parameter values:

$$r_c = 0.25\ \mu\text{m},\ R = 2.50\ \mu\text{m},\ \mu_{dr} = 1\cdot10^{-9}\ \text{m}^2/(\text{V}\cdot\text{s}),\ \Delta_e = 2.9\ \text{eV},\ \varepsilon = 3.$$

Computational results and discussion

Let us consider to what extent the shape of the charge distribution in the sample affects the dependences $F_c(t)$ and $F_{fr}(t)$. Fig. 2 shows the dependences calculated for U_{amp} = 1500 V and Δ = 150 ns using different approximations for the function $q(r,t)$.

The time variation in the field strength $F_c(t)$ at the cathode is shown in Fig. 2,*a*. Evidently, different approximations of the dependence $q(r,t)$ do not significantly affect the shape of the dependence $F_c(t)$. Three characteristic time intervals can be distinguished on all $F_c(t)$ curves, occurring due to two competing factors: an increase in voltage across the sample as described by Eq. (2), and the screening of the external field by the NSC near the cathode.

The first segment on the dependence $F_c(t)$ corresponds to a relatively low electric field strength, at which the influence of NSC on $F_c(t)$ is negligible. The increase in the electric field strength at the cathode during this time interval is associated only with a linear increase in the voltage across the sample and coincides with the dependence $F_c(t)$ in the absence of NSC (dashed line *4* in Fig. 2,*a*).

The second time interval on the dependence $F_c(t)$ begins when F_c reaches a critical value, at which the field emission current increases sharply (see dependence $j_c(t)$ in Fig. 2,*c*) and NSC accumulates rapidly near the cathode. Apparently, the screening of the external field by the NSC near the cathode occurs at a faster rate than the increase rate of F_c induced by the rising voltage. A maximum is formed on the $F_c(t)$ dependence. As NSC accumulates, its screening

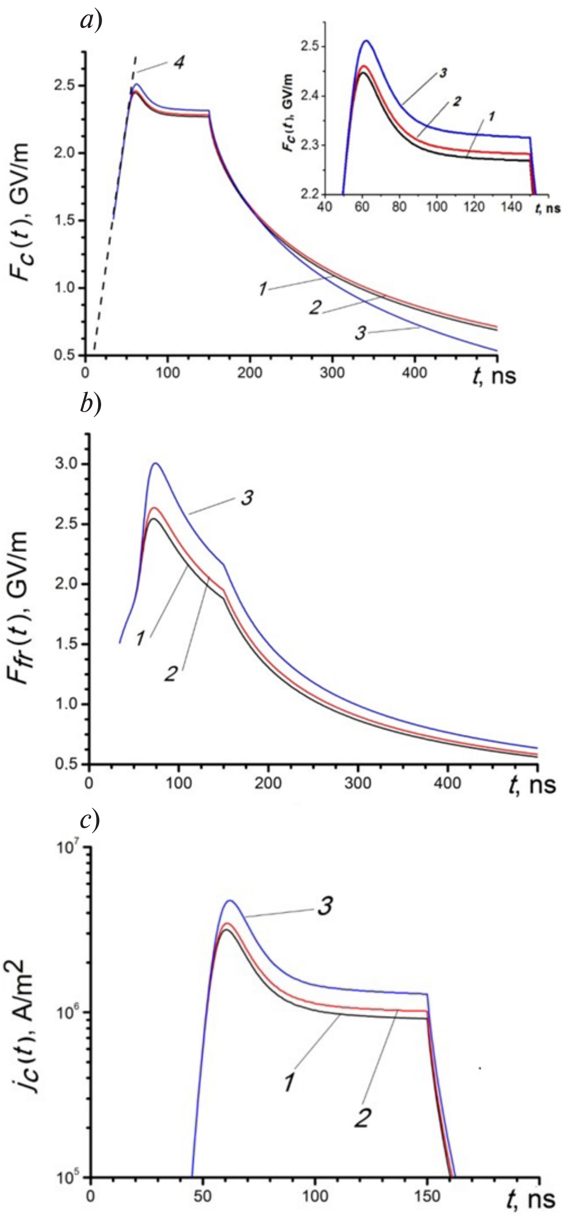

Fig. 2. Dependences $F_c(t)$ (*a*), $F_{fr}(t)$ (*b*), $j_c(t)$ (*c*), calculated using different functions approximating $q(r,t)$: quadratic (see Eq. (12), curve *1*), linear (14) (curve *2*); cubic (11) (curve *3*); dashed line *4* shows the variation in $F_c(t)$ in the absence of space charge screening
Inset: detailed image of the initial section of $F_c(t)$ curve

efficiency increases, and the value of F_c stabilizes. As a result, the current density $j_c(t)$ also stabilizes (see Fig. 2,*c*). A plateau is formed on the $F_c(t)$ and $j_c(t)$ dependences. The magnified inset in Fig. 2,*a* provides a clearer illustration of how the approximating function $q(r,t)$ influences the dependence $F_c(t)$ during NSC screening near the cathode. Apparently, the differences between curves *1*–*3* are insignificant, but the NSC limits F_c more efficiently if a second-degree polynomial is chosen as an approximating function (curve *1*, corresponding to a quadratic function).

The third time interval on the dependence $F_c(t)$ starts when the voltage across the sample stops increasing, i.e., at $t = \Delta$. A rapid decrease in field strength at the cathode is observed in the pulse plateau. Notably, by this point in time, the NSC layer penetrates into the polymer to a depth of 0.15–0.25 µm, so that the greatest penetration depth corresponds to the quadratic polynomial approximating the function $q(r,t)$. The reason for the decrease in F_c over time is that the voltage across the sample stops growing at $t \geq \Delta$ (pulse plateau), but NSC continues to accumulate despite a decrease in the injection current density (see Fig. 2,*c*). Consequently, the screening efficiency of the NSC increases and the values of F_c and j_c decrease, tending to their steady-state values.

The dependences $F_{fr}(t)$, constructed using various trial functions approximating $q(r,t)$, are shown in Fig. 2,*b*. Similar to the $F_c(t)$ dependences discussed above, they exhibit only minor discrepancies. We can therefore conclude that the calculated electric field distribution in the given electrode geometry is largely insensitive to the particular choice of the approximating function meeting the above criteria.

Analyzing the dependence $F_{fr}(t)$, we should bear in mind that the accumulating NSC enhances rather than limits the field strength at the boundary of the layer where it accumulates. However, the NSC zone keeps shifting deeper into the polymer. Consequently, the electric field component determined by the geometry of the electrode system is decreased, so the distinct stages observed on the $F_{fr}(t)$ curves are far less pronounced than on the corresponding $F_c(t)$ curves.

The values of F_{fr} and F_c are approximately the same for the same times t, i.e., the electric field distribution is close to uniform near the cathode where NSC accumulates. For F_{av} = 666 MV/m (which corresponds to U_{amp} = 1500 V) and a polymer film thickness of 2.25 µm, the electric field strength in the NSC layer reaches values of about 2.5 GV/m. Such a high-strength field exists on a nanosecond time scale within this region of the sample, approximately corresponding to the rise time of the electric pulse.

It is known that the steepness of the pulse's leading edge significantly affects many phenomena observed in polymer dielectrics in strong electric fields. For example, a sharp increase in the breakdown strength of the sample is observed for a PET film with an increase in the pulse front steepness [11], accompanied by an increase in electroluminescent brightness of this polymer [24]. According to the observations in [24, 25], electroluminescence only occurs in the polymer in the leading edge of the pulse and is absent during its plateau.

Let us consider the effect of the pulse's rise time (with U_{amp} = 1500 V) on the dependences $F_c(t)$ and $F_{fr}(t)$. Note that all calculated dependences here and below are obtained by approximating the function $q(r,t)$ by a quadratic polynomial (see Eq. (12)). The simulation results are shown in Fig. 3. Evidently, increasing the pulse front steepness (by decreasing its rise time at a constant amplitude) leads to an increase in the electric field strength both at the cathode (see Fig. 3,*a*) and at the boundary of the NSC accumulation zone (see Fig. 3, *b*). However, the plateau has a shorter duration on the dependences $F_c(t)$ and $F_{fr}(t)$, where the field strength reaches the highest values. For example, as the front steepness increases from 5 to 10 GV/s, the maximum field strength increases from 2.30 to 2.45 GV/m near the cathode (such an increase in F_c leads to an increase in the injection current density from 1.2 to 3.2 MA/m^2) and from 1.70 to 2.55 GV/m in the NSC accumulation zone. However, the duration of the plateau decreases from 190 to 100 ns on the $F_c(t)$ dependence and from 160 to 75 ns on the $F_{fr}(t)$ dependence.

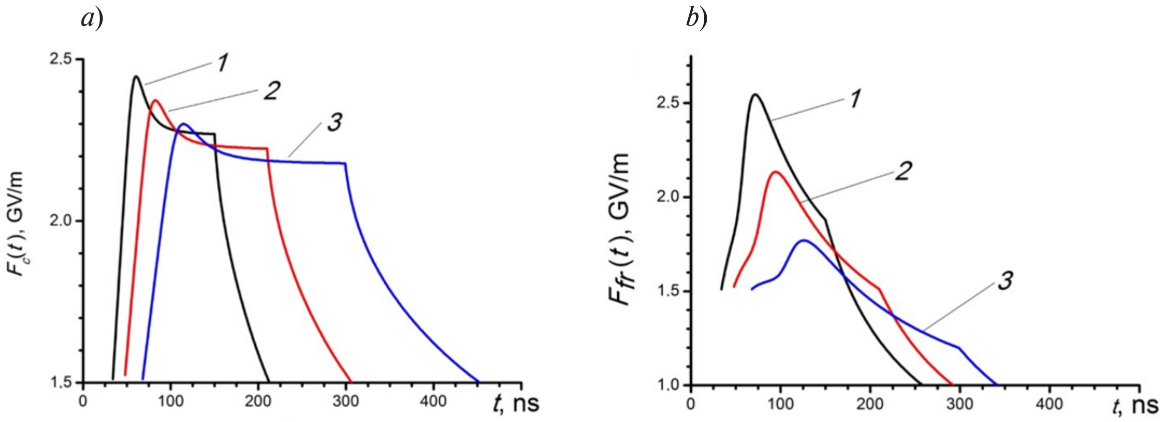

Fig. 3. Dependences $F_c(t)$ (a) and $F_{fr}(t)$ (*b*) calculated at U_{amp} = 1500 V and different pulse front steepnesses, GV/s: 10 (*1*), 7 (*2*) and 5 (*3*)

Conclusion

Analyzing the accumulation dynamics of the negative space charge (NSC) in a polymer exposed to high-voltage pulses (front steepness of about 1 GV/s) and the redistribution of the electric field *F* resulting from the application of these pulses, we can formulate the following conclusions. An NSC forming in the near-cathode region (within approximately 100 ns) simultaneously limits the electric field strength near the cathode and increases it at the boundary where the NSC penetrates into the polymer, producing a more uniform field distribution overall. The field strength *F* in this zone of the polymer dielectric, which is several times higher than its average over the sample, occurs only in the leading edge of the high-voltage pulse. A shorter rise time of the high-voltage pulse leads to an increase in the electric field strength of the polymer within the NSC accumulation zone while reducing the exposure time to this strong field. On the one hand, this combined effect may accelerate the field-initiated processes in the polymer material, but on the other hand, it shortens their duration.

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