Prebreakdown characteristics of weakly ionized liquid and gaseous media in the strongly nonuniform electric field

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Abstract. A theoretical model of electrohydrodynamic prebreakdown phenomena in slightly ionized (weakly conductive) media is proposed. The electric high voltage conduction of weakly conductive liquids and slightly ionized gases in intense electric fields using this model is considered. The formula for the calculations of volt–ampere characteristics under high voltage spherical capacitor field is analytically obtained. The experimental corona discharge volt–ampere characteristics of air are presented. It was found that the size of the ionization region in the case of corona discharge in air increases monotonically with increasing discharge voltage.

1. Introduction
The deviations from Ohm’s law for slightly ionized solid dielectrics such as mica in prebreakdown stationary fields of a plane capacitor were experimentally discovered by Poole [1].

Empiric stationary dependences of the electric current on an applied constant voltage \( I(U) \) were approximated by exponential dependences. For liquid weakly conductive media such as transformer oil, empirical dependences similar to those of Poole were obtained in [2]. For slightly ionized gases such empirical exponential dependences are known as Nikuradse’s curves. They are described in [3], alongside with similar curves for weakly conductive liquid dielectrics. In [4], the author analyzes the effect of the prebreakdown electroconvective transport that is formed in weakly conductive liquid dielectrics of the spatial charge on their conductivity and, hence, on such characteristics. For weak electrolytes in electrochemistry courses [5], this empirical effect of the exponential character in the field of an elongate plane capacitor is also referred to as the second effect of Wien (the first effect of Wien in electrochemistry is considered to be a similar but a much weaker effect for strong electrolytes, which is caused by the effect of the prebreakdown electric field not on the concentrations but on the mobility of ions). In a strongly ionized plasma [6], the effect both of uniform and nonuniform electric fields on the nonlinearity of the volt–ampere characteristics is approximately the same as in strong electrolytes. In drastically nonuniform prebreakdown fields both in weakly conductive liquids and slightly ionized gases [7], the most typical among the observed prebreakdown deviations of Ohm’s linear law are quadratic.

Theoretically, the exponential growth in conductivity of slightly ionized solid media of a semiconductor type with an increase in the modulus of intensity of the electric field up to the magnitude of electric breakdown was justified by Frenkel [8]. In [8], an equation of the Arrhenius
type was used for the dependence of the volume ionization rate of such media on temperature, taking into account a decrease in the potential of ionization of their neutral particles. It is noteworthy that, e.g., in [7], the fairly simple, almost identical algebraic Schottky estimates performed earlier were described. They were carried out for a similar decrease in current efficiency (owing to the prebreakdown electric field) during thermal emission of electrons from a heavily heated high voltage cathode. For the dissociation constant of weakly conductive liquid dielectrics in [9], its dependence on the modulus of the intensity of the electric field in the form of the Frenkel exponent was derived, taking into account the decrease in the activation energy of their molecules that are partially dissociated. In [10], using the methods of physical kinetics, the author obtained a dependence of the dissociation constant of weak electrolytes an analytical and zero Bessel’s function on the modulus of electric field intensity of the first order of an imaginary argument (unlike the Frenkel exponent, which grows, according to [11], almost in the same way under prebreakdown fields).

Therefore, in electrochemistry [5] it is commonly believed that Onsager theoretically justified the empirical effect of Wien of the prebreakdown nonlinearity of the volt–ampere characteristics for weak electrolytes. Here, as in [8] and [10], the transition from the dependence of the ionization rate (dissociation) to the dependence of conductivity on temperature and the modulus of electric field intensity was carried out using the law of active masses (the proximity of the volume rate of ionization (dissociation) of neutral particles and recombination of the charged ones). It was carried out on the assumption of the chemical (dissociative) or ionization equilibrium of media without taking into account (beyond establishing influence) the nonstationary change in the low-voltage conductivity occurring in them, up to almost prebreakdown conductivity. Also the effect of the diffusion of the charged particles on it and their drift rate in strong electric fields were not taken into account. Furthermore, we do not take into account the effect of the flows of the liquid and gaseous media, mentioned here. These flows arise in such fields due to the formation of prebreakdown volumetric charge in them on the spatial distribution of the density of such a charge, which, as is shown in [4], can also change the spatial and temporal distribution of conductivity.

In [11–13], macroscopic equations were derived, taking into account the effect in a general case of these processes on the nonlinearity of the volt–ampere prebreakdown characteristics of weakly conductive liquid dielectrics under study.

In [11, 13], the possibility of applying the formulas of Frenkel and Onsager was studied theoretically. It was shown that for their use in the case of the validity of Longevin–Onsager formulas [10] for the dependence of the coefficient of recombination of the charged particles on their mobilities, it is sufficient to fulfill the condition of the quasi-neutrality of the weakly conductive liquids under study with ion conductance not only in weak but also in prebreakdown electric fields. For the classic electron–ion plasma, similar near-electrode layers of violation of quasi-neutrality are known as the Langmuir layers [6], and for strong electrolytes as the Debye layers [5].

In the case of the isothermal regimes implemented for given media in the absence of external heating and for weak Joule heating by the field current of the plane high-voltage capacitor, Frenkel exponential dependence of volt–ampere stationary characteristics on the applied constant voltage theoretically follows from the algebraic equation of the stationary law of charge conservation in a differential form. In the case of a spherical capacitor from this law we derived in [13] an ordinary differential equation of the first order for determining the stationary distribution of the electric field potential in its interelectrode gap, which is filled with a weakly conductive liquid such as transformer oil. This equation describes the change in the potential of a spherically symmetric electric field along the radial coordinate.

In [13], the analytic solution of this differential equation was also obtained. Under the condition of the equality of the modulus of the potential difference between the capacitor
electrodes to the applied constant voltage, it is followed by the expected ohmic linearity of the volt–ampere characteristics in the weak fields and by its quadraticity in the prebreakdown ones for large interelectrode gaps. This theoretic result also agrees well with the prebreakdown empiric curves for weakly conductive liquid dielectrics that were obtained experimentally by different authors. However, in the case of corona discharge in slightly ionized gases [3], microlevel processes differ markedly from those of weakly conductive liquid dielectrics. Nevertheless, the quadratic dependence for the gases of the type of the quadratic dependence that we obtained fairly long ago for weakly conductive liquid dielectrics [13] was derived by Townsend [7], though not in the field of spherical but rather elongate cylindrical condensers with a filamentous internal corona-forming electrode. The Townsend quadratic dependence of $I(U)$ for gases is also frequently supported experimentally, with the results described in [7].

With a decrease in the interelectrode gap even for nearly high-voltage point electrodes for weakly conductive liquid dielectrics this characteristic ceases to be quadratic, and its growth becomes more abrupt, sufficiently close to the exponential growth of the prebreakdown field of plane elongate capacitor. Therefore, this characteristic can be considered quasi-exponential, that is, for the given liquids, intermediate between exponential dependence in the field of the plane elongate capacitor and quadratic dependence in the field of close to point or filamentous high voltage electrodes (with counter electrodes of sufficiently large sizes compared to them, removed far from them), by nature of its monotonous growth. But we did not carry out quantitative calculations in [13] of this transcendental prebreakdown volt–ampere characteristic.

In addition, the results of our experiments [14, 15] are evidence that for slightly ionized gases at not very close interelectrode gaps between corona-forming electrodes and counter electrodes with larger sizes, both Townsend quadraticity and increasing deviations from it with $U$ growth are observed. Hence, our study is aimed at obtaining, using our theoretical model (not appropriate for gases according to [3]), analytical formulas for the abovementioned quasi-exponential prebreakdown dependences in weakly conductive liquid dielectrics. Another aim of the work is to compare the results of our experiments [14, 15] with dry air in the field of the corona-forming electrodes with the calculation results of Townsend and Tikhodeev, which differ from our calculations for the weakly conductive liquid dielectrics. The results of this comparison additionally prove the presence of deviations from the more traditional prebreakdown quadraticity of volt–ampere characteristics of a strongly nonuniform field for the complication of the geometry of both corona-forming electrodes and counter electrodes.

2. The calculation formula for the quasiexponential volt–ampere characteristic of a spherical capacitor filled with weakly conductive liquid dielectric

At obtaining analytical calculation quasiexponential dependence for the prebreakdown nonlinear volt–ampere characteristics of weakly conductive viscous liquid dielectrics such as transformer oil, we used the same theoretical model as in our earlier works [11, 13]. According to [9], even impurity free liquids of this kind are considered to be analogues of weak electrolytes. Some components of the rings of their molecules of a benzene type with a complex composition with ionic bonds can dissociate in the absence of an electric field applied to such media due to the thermal motion of these molecules (described in classics) and their collisions because of this motion. This dissociation appears to be very weak. In the interaction of weakly conductive liquid media, both purified and unpurified, with electric field applied, strengthening occurs as in weak electrolytes according to Onsager [10]. It becomes far more intense (exponentially so, in accordance with Frenkel and Plumley [8, 9]) with increases in the modulus of intensity of the field but of a lesser breakdown force. Their ohmic conductance

$$\sigma = (n⁺b⁺ + n⁻b⁻)Ze$$

(1)
grows, but prior to the electric breakdown the composition of neutral components, it varies slightly. In (1), $n$ are the volume concentrations of ions with opposite signs; $b$ are the values of
their electrical mobility; \( Z \) is their valence or the repetition factor of the bound ion pairs such as HO (the case of purified transformer oil [16]) located along the edges of benzene rings of the molecules and partially dissociating during collisions. Here, the nucleus of the benzene rings of molecules of CH type (also the case of purified transformer oil [16]) remains neutral until the electric breakdown of the medium. This statement can obviously be strictly justified by the methods of quantum chemistry with the determination of value \( Z \) in (1) for each case of the weakly conductive liquid under study.

In addition, in (1) \( e \) is the value of the elementary charge (either proton or electron). In the case of a spherical high voltage capacitor with the studied liquid dielectric layer with thickness \( d \) in the interelectrode gap, we use a generalization in the case of different values of mobilities of positive and negative ions in the system derived in [13] of stationary electrodynamic equations of a diffusionless one-dimensional approximation. This generalized system of equations, which we have derived on the analogy of the one derived in [13] without taking into account the influence of the prebreakdown flows of media on the sought nonlinearity of their volt–ampere characteristics, is as follows:

\[
\sigma E = \frac{I \text{sgn}[\varphi^\pm(r_0)]}{4\pi r^2}, \quad E = -\frac{d\varphi}{dr};
\]

\[
(\varepsilon\varepsilon_0)^2 b_+ b_- E \Delta, E = \sigma_0^2 \exp(\beta \sqrt{|E|}) - \sigma^2 + (b_+ - b_-)\varepsilon\varepsilon_0 \left(\frac{dE}{dr} + \frac{2E}{r}\right)\sigma; \quad (2)
\]

\[
\beta = \frac{(Ze)^{3/2}}{(\pi\varepsilon\varepsilon_0)^{1/2}k_B T}, \quad U = \left| \int_{r_0}^{r_{0+d}} Edr \right|.
\]

In (2), the sign of the potential of \( \varphi(r_0) \) on the internal electrode in the considered stationary case is similar to the sign of its charge, \( \varepsilon \) is the dielectric permeability of the media under study, and \( \varepsilon_0 \) is the dielectric constant; \( k \) is the Boltzmann constant, and the absolute temperature \( T \) for the modes under study can be considered constant and room temperature.

In [13], a system of one-dimensional stationary equations analogous to (2) has been written both for the spherical and elongate cylindrical capacitors taking diffusion into account, but as was mentioned above, for the case of equal values of mobilities of oppositely charged ions. In both systems, we used a nonanalytic in zero exponential dependence of the dissociation constant of weakly conductive liquid dielectrics on the root from the modulus of the electric field intensity according to [8], together with a linear Langevin dependence of recombination coefficient on ionic mobilities. Its derivation is described in [7, 10]; (in [10], along with the derivation of the Langevin formula using the methods of physical kinetics, it is also shown that the recombination coefficient is independent of the modulus of the electric field intensity). In (2) \( \Delta, E \) is the Laplacian component of the vector function in a curvilinear system of spherical coordinates along its radial axis (in the case of spherical symmetry). The center of this coordinate system is located in the center of the internal electrode of a spherical high-voltage capacitor. In this situation, changes in characteristics for its angular coordinates are considered negligibly small compared to those along the radial ones. The boundary conditions in the one-dimensional stationary case, according to [9, 12], are as follows:

\[
n^A_{+} = n^K_{-} = 0. \quad (3)
\]

These conditions are caused by the repulsion of liquid ions from the electrode whose sign is opposite to the signs of the ions. From equation (3), the first equation from (2), which is the law of conservation of charge in a differential form, we can easily derive equations (1) in the dependence of \( \sigma \) on \( n_\pm \) of the electrodynamic equation of Gauss (\( \text{div} E = q/|\varepsilon\varepsilon_0| \)), and the equation of the dependence of density of the volumetric charge on \( n_\pm (q = |n_+ - n_-|Ze) \), the stationary boundary conditions for the intensity at the plates of a spherical capacitor. For the
derivation of these conditions, the differential one-dimensional Gaussian equation in a spherically symmetric case (one of the one-dimensional cases) is replaced by the difference equation. In (3), the upper indices denote the anode and cathode, relatively. In [9], the boundary conditions of (3) were shown, and in [12] were mathematically justified for the plane capacitor.

In [13], we generalized them in the case of electrophysical (electrochemical) processes of the neutralization of ions and the ionization of neutral particles of the weakly conductive liquid dielectrics under study. The latter type of electrochemical or ionization reactions for the electrode according to [3, 16] can lead to emission (injection) currents from the electrodes in strong fields. But in this work as well as in monography [17], the cases of injection or emission of charges from high voltage electrodes in a weakly conductive liquid dielectric are not regarded. In equations (2), \( \varphi \) is the potential of the electric field, whose distribution in a spherically symmetric case depends only on the radial coordinate \( r \): \( r_0 \leq r \leq r_0 + d \), where the thickness of the layer of the liquid dielectric (which fills the capacitor) dis the interelectrode gap. In addition to the previous designations in (2), \( \sigma_0 \) is the low voltage conductivity of such weakly conductive liquid dielectrics. The mobilities in them can be considered independent of the field. By analogy with our earlier work [13] and for different values of mobilities from (2) the prebreakdown condition of quasi-neutrality, the results are as follows:

\[
1 \gg a = \frac{\varepsilon \varepsilon_0 b_s}{\sigma_0 d^2 \exp \left[ \beta \left( U/d \right)^{1/2} / 2 \right]}, \tag{4}
\]

Under this condition, the solution of the stationary external problem of the equation system (2), which according to [13] (upon the Langevin dependence of the recombination coefficient on the ionic mobilities), corresponds to the chemical equilibrium of the media under study, is reduced to the solution of a far more simple equation system with a region of determination of the sought alternatives outside the boundary layers of violation of such quasi-neutrality (in (4) \( b_s = (b_+ + b_-)/2 \)). Such a system appears as follows:

\[
\begin{align*}
\sigma E &= \frac{I \text{sgn}[\varphi^\pm(r_0)]}{4\pi r^2}, \quad E = -\frac{d\varphi}{dr}; \\
\sigma &= \sigma_0 \exp \left( \frac{\beta E}{2} \right)^{1/2}; \\
\beta &= \frac{(Ze)^{3/2}}{\left( \pi \varepsilon \varepsilon_0 \right)^{1/2} k_B T}, \quad U = \left| \int_{r_0}^{r_0+d} E dr \right|.
\end{align*} \tag{5}
\]

The sign of the charge of the internal electrode, the same as the \( \text{sgn}[\varphi (r_0)] \), is either negative or positive in the stationary case under study, whereas the sign of the forming volume charge outside the near-electrode layers of violation of quasi-neutrality coincides with the sign of the charge of the internal electrode. The latter follows from (5). Namely, the differentiation of an implicitly preset function of \( E(r) \) leads to the following function for the volume density of the spatial charge:

\[
\begin{align*}
q &= \frac{\varepsilon \varepsilon_0 \beta E |E|^{1/2}}{r(1 + \beta E^{1/2}/2)}, \quad \beta = \frac{(Ze)^{3/2}}{\left( \pi \varepsilon \varepsilon_0 \right)^{1/2} k_B T}; \\
E &= -\frac{d\varphi}{dr}, \quad U = \left| \int_{r_0}^{r_0+d} E dr \right|; \\
E \exp \left( \frac{\beta E}{2} \right)^{1/2} &= \frac{I \text{sgn} \varphi(r)}{4\pi \sigma_0 r^2}.
\end{align*} \tag{6}
\]

From (6), it follows that the sign of a monotonically decreasing function of the volume charge density \( q(r) \), which substantially changes the applied field, coincides with the sign of the charge.
of the internal electrode. The third equation of (5) is a generalization in case of \( Z > 1 \) of Frenkel’s formula for conductivity from [8]. This generalization was proposed by Ostroumov in [17] for the first time. From this, the expression for the density of the steady volume charge (6) can be also obtained using another expressions from [17]:

\[
q = -\varepsilon_{0}(E \cdot \nabla \sigma)/\sigma
\]

provided that \( \sigma \) is defined according to (5). From equations (5), to determine \( E \) and \( U \), which we have derived, taking into account the mathematical justification of the neglect of the potential decrease in non-quasi-neutral boundary layers carried out in [18] by the method of asymptotic boundary layer disintegrations [19] with \( \alpha \ll 1 \), we obtain for the spherical capacitor by analytical integration and limiting transition close to the point high-voltage internal electrode \( \beta E_{1}/2 \gg 1 \), and at finite \( d \) we have:

\[
\tau_{d} = \frac{\varepsilon\varepsilon_{0}}{\sigma_{0} \exp (\beta E_{d}^{1/2}/2)};
\]

\[
I = \frac{16(\varepsilon Z)^{3}U^{2}}{\tau_{d}(256 + 64\beta E_{d}^{1/2} + \beta^{2}E_{d})(k_{B}T)^{2}};
\]

\[
r_{0} \leq r \leq r_{0} + d;
\]

\[
E_{0} = |E(r_{0})|, \quad E_{d} = |E(r_{0} + d)|.
\]

The quadratic volt–ampere characteristic (that we obtained in [13]) at \( \beta E_{0}^{1/2} \gg 1 \) and \( d \rightarrow \infty \) can also be obtained from [7] at \( d \rightarrow \infty \). Namely:

\[
I = \frac{(\varepsilon Z)^{3}U^{2}}{16\tau_{0}(k_{B}T)}.
\]

Here, together with the aforementioned, \( \tau_{0} = \tau_{d \rightarrow \infty} = \varepsilon\varepsilon_{0}/\sigma_{0} \).

In the prebreakdown electrohydrodynamics of weakly conductive liquids, \( \tau \) as the time of relaxation of forming prebreakdown volume charge in them was used earlier [20, 21]. For liquid dielectrics such as transformer oil, it is of an order of 1 second even in weak fields (according to [16, 22]). To round off the prebreakdown volt–ampere characteristics of weakly conductive liquid dielectrics such as transformer oil, we must note that from the formula (8), which we have obtained theoretically, the growth of the prebreakdown current occurs at fixed \( U \) with a decrease in \( d \). This is easy to see, since the sign of the derivative of the implicit function (8), which is relevant to the dependence \( I(d) \), with a fixed \( U \) and distribution \( E(r) \) definable by the use of (6), is negative. In addition, we should note the substantial effect of the molecular composition on the current value at fixed voltage. Namely, monotonously increasing dependence of \( I(Z) \) for a constant \( U \) also follows from formula (8), which we have obtained, as well as form (9), in which this dependence is cubic.

### 3. Empiric dependence of prebreakdown characteristic in the case of the corona discharge in slightly ionized gases at corona-forming pinpoint

The slightly ionized gases, like air, called electronegative [7] are closest in molecular composition and to the type of the prebreakdown dependence \( I(U) \), particularly in strongly nonuniform fields, which were described in the previous section, about a weakly conductive liquid dielectric, because of the partial adhesion of their electrons to neutral molecules. Upon the interaction of such media with a strongly nonuniform field of high-voltage electrodes with a type of pin, blade, high voltage wires, or small-size spheres, a corona glow is observed [7] near these electrodes. In such boundary-layer regions, the electrodynamic characteristics depend on free electrons as well as on positive and negative ions. In the field of a high-voltage corona-forming cathode the
electrons can enter the gas not only through radiation, as they do in weak fields, but also due to the different types of electron emission that are thoroughly described in [7]. Ions in such gases for the applied prebreakdown fields are formed not only due to the sticking of electrons to molecules, but also due to the shock ionization of the molecules by the free electrons that move in strong fields. In addition, they are formed owing to chemical processes of plasma, which differ from the dissociative in weakly conductive dielectrics that were described in the previous section.

Alongside with the internal zones of discharge adjacent to the corona-forming electrodes, there are also external regions with unipolar charging in the stationary cases [7, 20, 23]. In them, the sign of the forming volume charge upon its establishment coincides with the sign of the charge of the corona-forming electrode (analogous but quasi-neutral zones for the weakly conductive liquid dielectrics that we have already described above).

4. Experimental results of prebreakdown characteristic in the case of the corona discharge in air

The experiments were carried out in air at atmospheric pressure. The needle with an edge radius equal to 70 µm was placed above the center of a flat round electrode with a diameter of 100 mm at a distance of 13 mm. In this case, the finite size of the flat electrode can be neglected. High dc voltage of negative polarity was applied to the needle. The flat electrode was grounded. The dc voltage was measured using voltage divider with the voltage ratio of 1 : 1000 and multimeter APPA 505. The error of measuring the dc voltage at the limit of 10 V was equal to ±0.17 V. The current was measured using ammeter with the accuracy class of 0.5.

We believe that the corona discharge region where the ionization processes occur corresponds to the region emitting in the visible range. Photography was used to determine the size of this area.

Figure 1. The volt–ampere characteristics of the corona discharge of negative (circles) and positive (crosses) polarity.
Figure 2. Photos of the discharge with the following parameters: (a) $I = 6 \, \mu A$, $U = 5.01 \, kV$; (b) $I = 27 \, \mu A$, $U = 8.01 \, kV$; (c) $I = 68 \, \mu A$, $U = 11.04 \, kV$.

Figure 3. The distance from the end of the needle to the boundary of the ionization region along the vertical axis (circles) in comparison with the square root of the cross-sectional area (crosses) of the ionization region in the case of the corona discharge in air.

Nikon 1 V2 (4608 $\times$ 3072) camera with Nikon 60mm f/2.8D AF Micro-Nikkor lens was used for taking pictures. The photography was carried out with a fully open aperture, the exposure was equal to 5 s, and the photosensitivity was equal to 400 ISO units. A pixel of the matrix corresponded to a 0.0044 $\times$ 0.0044 mm$^2$. The position of the camera during the experiment did not change.

Figure 3 shows size of the ionization region in the case of the corona discharge in air. Figure 1 shows the volt–ampere characteristics of the discharge. One can see that in the case of the corona discharge in slightly ionized gases like air volt–ampere characteristics for
positive and negative discharge are significantly different in contrast to weakly conductive liquid dielectrics.

Figure 2 shows photographs of the discharge for various parameters. At the discharge voltage less than 7 kV the glow area is stationary and vertically symmetrical and expands with increasing discharge current. At higher discharge currents, the glow area loses symmetry and begins to fluctuate. The breakdown occurred at the discharge voltage more than 12 kV.

It can be seen that the size of the ionization region increases monotonically with increasing discharge voltage.

5. Conclusion
This study presents in the form of graphs and formulas the monotonously growing dependences of the prebreakdown stationary volt–ampere characteristics of weakly conductive liquid dielectrics as well as slightly ionized gases. Obtained characteristics differ from the traditional volt–ampere characteristics for point electrodes by a sharper current increase with increasing applied voltage.

In the case of the interelectrode gap of the high-voltage spherical capacitor being filled with viscous heat-conducting liquid dielectrics such as transformer oil independent of the change in polarity of the electrodes, the calculation formula testifies to the growth of the prebreakdown current with a decrease in the interelectrode gap. Such a formula can be used for calculations of spatial distributions of stationary rates of developed flows of weakly conductive liquid dielectrics from thin high-voltage axis-symmetrical electrodes considering the condition of adhesion on the counterelectrode.

It was found that in the case of the corona discharge in slightly ionized gases like air volt–ampere characteristics for positive and negative discharge are significantly different in contrast to weakly conductive liquid dielectrics. The size of the ionization region in the case of corona discharge in air increases monotonically with increasing discharge voltage.

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