AN ASYMMETRICAL DIELECTRIC BARRIER DISCHARGE IN THE PULSED MODE

D.G. Korenyugin[1], A.M. Martsinovsky[2]

[1] Peter the Great St. Petersburg Polytechnic University, St. Petersburg, Russian Federation;

[2] The Ioffe Institute of the Russian Academy of Sciences, St. Petersburg, Russian Federation

The paper presents the results of an investigation of an asymmetric dielectric barrier discharge (DBD) in the air at atmospheric pressure. The discharge system consisted of a plane electrode and semispherical short-radius one, and the both were coated with dielectric. The discharge was excited by a train of almost sinusoidal damped oscillations which was applied to electrodes with a frequency of 1 kHz. It has been found that the preferable to biological applications homogeneous avalanche form of the DBD is always realized in the investigated voltage range 7 – 15 kV, whereas the streamer form of the discharge is excited under identical conditions in the case of a conducting semisphere. It was also established that the positive and negative (according to the voltage sign of the first pulse of the train onto the small electrode) discharge differed greatly in their characteristics. An explanation for this difference has been proposed.

Key words: dielectric barrier discharge; pulse train; avalanche; streamer form; discharge mode

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Introduction

A barrier discharge is a gas discharge occurring in the gap between electrodes, where at least one of the electrodes is coated with a dielectric. Such discharges are traditionally used for various purposes, and, as a rule, their discharge systems have a plane-parallel electrode configuration. If the interelectrode gap is narrow enough (on the order of millimeters), the distribution of the electric field in the discharge region can be considered homogeneous, so this type of barrier discharge is symmetrical, and this subject has been well understood. However, more complex asymmetrical barrier discharges with an inhomogeneous electric field distribution are being actively studied for some practical applications, primarily, plasma technologies in medicine. The discharge systems of such barrier discharges are usually cells where one of the electrodes is flat and the other can have an arbitrary shape. The most frequently used electrode systems are the sphere–plane and the tip–plane types.

The main form that an asymmetrical barrier discharge takes in air under atmospheric pressure is a coronal streamer [1, 2]; this is the form typically involved for practical applications. However, sometimes this type of discharge should not be used, in particular, in medicine and biology, when treating (sterilizing) relatively large open wounds or bacterial samples for a short time with a discharge [3 – 5]. The streamer form is insufficiently homogeneous for these purposes, and streamers can actually damage living tissues. In view of this, finding a more stable form of homogeneous avalanche discharge is of great interest. For example, according to the data in [6], a homogeneous form of an asymmetrical discharge in air, excited by high-voltage trains, evolved in the pulse frequency range $f \approx 10$ MHz with an interelectrode gap $L \leq 0.4$ cm. However, the discharge always changed into streamer form at lower pulse frequencies and larger gaps (which substantially simplifies practical applications). For this reason, searching for modes of a homogeneous asymmetrical barrier discharge that are convenient for practical use is an important problem, as is exploring the conditions (and causes) for the existence of the avalanche and streamer forms.

The goal of this study was to find the optimal modes of an asymmetrical barrier discharge with a homogeneous form.

This work continues the study of this type of discharge started in 2008 for a discharge system with a single dielectric electrode [2, 7]. In accordance with our main goal, we have set the task of examining a system with two dielectric electrodes and comparing the characteristics and patterns of the discharge for the two cases.

Experimental procedures

An asymmetrical barrier discharge in air under atmospheric pressure was studied in a discharge cell (Fig. 1). While previous studies [2, 7] used a metal or liquid-phase (conducting) electrode as the small electrode above the surface of this cell, in this experiment, a non-conductive polymer coating was applied to the surface of a spherical metal electrode *2* of radius r = 0.3 cm (cellulose acetate butyrate CAB-308) and 0.075 cm thick. The second electrode was a 0.25 cm thick glass plate *1* with a translucent conductive coating applied to its lower surface. The distance from the sphere to the plate was L = 0.25 cm. A camera capturing images of the discharge from the end-face was placed below the plate.

Fig. 1, *b* shows a typical distribution of the electric field along the shortest distance from the sphere to the plate (in the interelectrode gap) for the voltages used in the experiment. Of course, the field is extremely inhomogeneous, so in general the field strength E_{br} for static breakdown ($E_{br} \approx 30$ kV/cm for air [8]) is reached in the interelectrode gap at a distance z_{ed} from the surface of the spherical electrode.

A sequence of high-voltage damped trains with oppositely polarized almost sinusoidal voltages was applied to the electrodes of the discharge system (see Fig. 3); the parameters of these voltages are given in the table. The pulse voltage generator provided train sequences with both $U_1 > 0$ and $U_1 < 0$ values relative to the grounded non-planar electrode.

The voltage across the discharge gap was measured with a calibrated antenna connected to an oscilloscope; the low-voltage values of

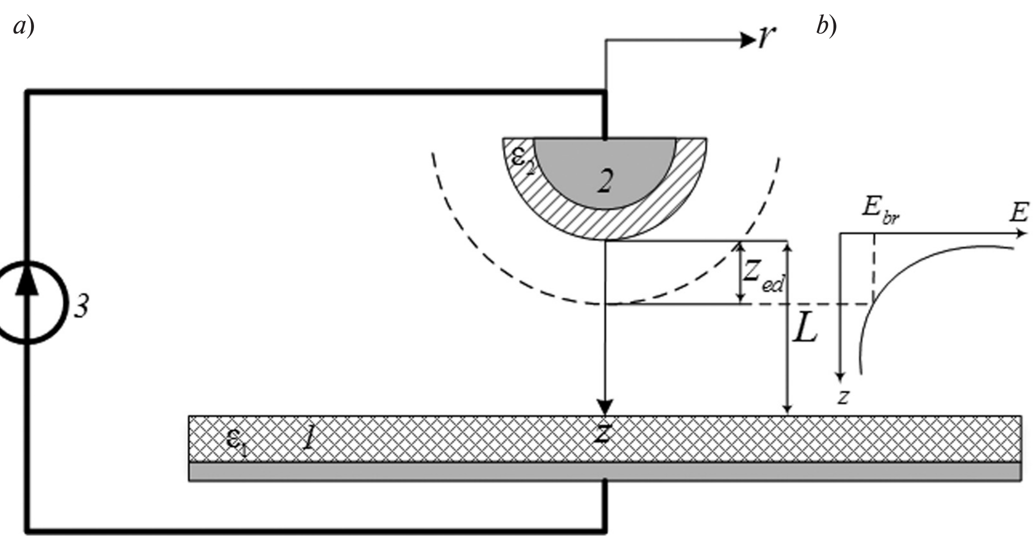

Fig. 1. Schematic of the discharge cell (*a*) and the electric field distribution in the interelectrode gap (*b*): flat glass plate *1* with conductive coating, spherical metal electrode *2* with non-conductive polymer coating, source *3* of high-voltage damped trains with oppositely polarized almost sinusoidal voltages; L, z_{ed}, E_{br} are the characteristic parameters of the barrier discharge; z, r is the coordinate system; E is the electric field strength; ε_1, ε_2 are the dielectric permittivities of electrode coatings

Table

Parameter values of high voltages applied to the electrodes

Parameter	Notation	Unit	Value
Train duration	τ_t	μs	≈150
Train repetition rate	T_t	ms	1
Oscillation frequency of pulses in a train	f	kHz	55
Pulse duration			
first pulse	τ_1	μs	4
subsequent pulses	τ_k		8 – 9
Voltage of first pulse	U_1	kV	7 – 15
Pulse damping	$U_1 : U_2 : U_3$	–	1.0 : 0.8 : 0.7

the antenna signal were then converted to high-voltage (using the calibration curve). The voltage could be additionally adjusted with the volts/div knob, usually with sufficient accuracy (±0.2 kV). The discharge current was measured with a 5-Ω non-inductive resistive shunt connected between the spherical electrode and the ground. The setup is described in more detail in [2, 7].

Experimental results and discussion

We have discovered in the course of the experiment that while using a polymer coating on the spherical electrode eliminated charge flow from the discharge gap to the external circuit and further impeded avalanche current (as the bias current had to be maintained in this dielectric layer), this also radically changed the discharge shape. Only a streamer form of an asymmetrical discharge could be generated in the experiments described in [2, 7] for a metallic or liquid conductive semi-sphere, virtually throughout the entire range of its parameters; however, a homogeneous avalanche shape of an asymmetrical discharge was steadily observed in a sphere with a dielectric coating with a wide range of pulse voltages from 7 to 15 kV for both polarities.

Fig. 2 shows typical integral images for the shapes of discharges with opposite polarities for a coated semisphere and for a semispherical surface of a droplet of a conductive liquid. Fig. 2, *a* shows the image obtained for a negative barrier discharge, i.e., for the case when a negative voltage was applied to the sphere and a positive voltage was applied to the plate.

Fig. 2, *b* shows the image obtained for a positive barrier discharge, i.e., with the reverse polarity voltage applied. Fig. 2, *c* shows the image for the experiment described in [2, 7] for the case of a negative asymmetrical barrier discharge. The voltage U_1 of the first pulse on the drop was about 13 kV. In the latter case, the discharge existed in a streamer form.

We should note that the polarity of the corona discharge (positive and negative corona) is typically determined by the sign of the voltage in the tip (the electrode with a small area), since the processes near it play a key role and depend on the voltage sign. The same is largely true for an asymmetrical barrier discharge. Therefore, it is logical to follow the same principle in determining the positive and negative asymmetrical barrier discharges by the sign of the voltage on the sphere relative to the plane electrode. In our experimental conditions, a voltage was applied as a train of oppositely polarized pulses, and it was assumed that the polarity was determined by the most intense first pulse, during which avalanche breakdown develops most often.

Observing the discharge from the side, we have established that it is a luminous cone from the semisphere to the plate, with an angle of about 30° for a negative discharge and about 15° for a positive one. In the first case, bright glow

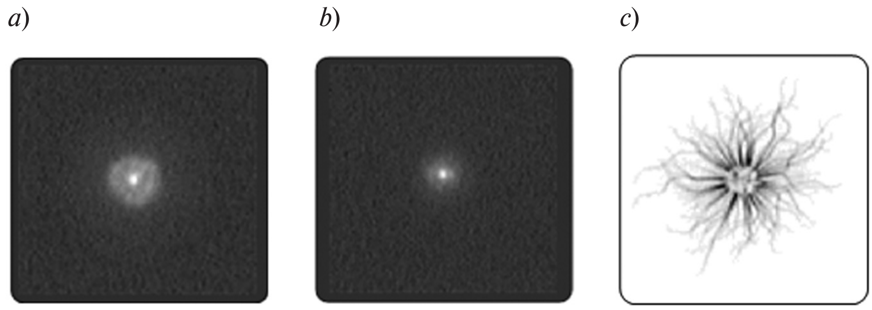

Fig. 2. Comparison of integral images of the shapes of oppositely polarized discharges for a coated semisphere (*a*, *b*) and for a semispherical surface of a droplet of a conductive liquid (*c*). The images shown are for the negative (*a*, *c*) and the positive (*b*) asymmetrical barrier discharges. The characteristic geometric parameters of the discharge system are the same in all cases.

propagated up to the surface of the plate, while in the second case the glow became noticeably dimmer towardss the plate and increasing the interelectrode gap leads to the glow "breaking away" from the surface of the plate.

The observed picture of the discharges clearly indicates that the field strength is high enough to not only ensure the propagation of avalanches but also initiate them in the entire discharge gap for the given experimental conditions; respectively, the distribution profile of the electric field, shown in Fig. 1,*b*, should apparently decrease to E_{br} more smoothly.

We should also note that the avalanche is actually a quasi-homogeneous or an integrally homogeneous form of discharge. Indeed, a lot of avalanches manage to develop during the exposure time (≈ 20 ms) with a train repetition rate of 1 kHz, and their total glow generates a homogeneous picture registered by the detector. However, from a practical point of view, even this form of discharge fully satisfies all the necessary requirements (for example, when used for rapid sterilization).

Our studies have revealed that while discharges of opposite polarities are close in form and type of glow, the conditions in which these discharges form turn out to be different.

Fig. 3 shows the electrical characteristics of a negative asymmetrical barrier discharge (the waveforms of the voltage $U(t)$ and pulse current $I(t)$ with different sweep speeds) typical of the range of voltages U_1 from 7 to 12 kV. Some explanation is required for the waveforms obtained. Since the spherical electrode was grounded in the circuit used, the oscilloscope recorded the voltage on the plate relative to this electrode (the ground). To determine the voltage on the spherical electrode itself relative to the plate, the polarity of the voltages and currents on the waveforms had to be changed.

Thus, it can be seen from Fig. 3 that on a millisecond scale steady discharge current is a sequence of individual short pulses of opposite polarities, differing in amplitude by an order of magnitude. Not every pulse train is accompanied by a current pulse.

The waveform with a nanosecond sweep of individual pulses (see Fig. 3, *d*) has the form typical for avalanche discharge [9]. This result, along with the range of maximum pulse currents (1 – 40 mA), also typical for avalanches, conclusively proves that it is the avalanche (Townsend) discharge form we are dealing with. Most avalanches are formed during the first pulses of the train, which are the most intense; in this case, either a pair of current pulses (first with a negative, then with a positive polarity, see Fig. 3, *b*), or one pulse of the corresponding polarity (Fig. 3, *c*) is observed.

In a negative discharge, avalanches move from the spherical electrode to the plate, into the region with the decreasing field. However, as we have pointed out above, the character of the discharge glow indicates that almost all of the avalanches reach the dielectric surface of the electrode in the given conditions, depositing a negative electron charge on it. This charge generates an electric field inverse to the external field, which prevents the formation of new avalanches, even when breakdown occurs before the high-voltage pulse reaches a maxi-

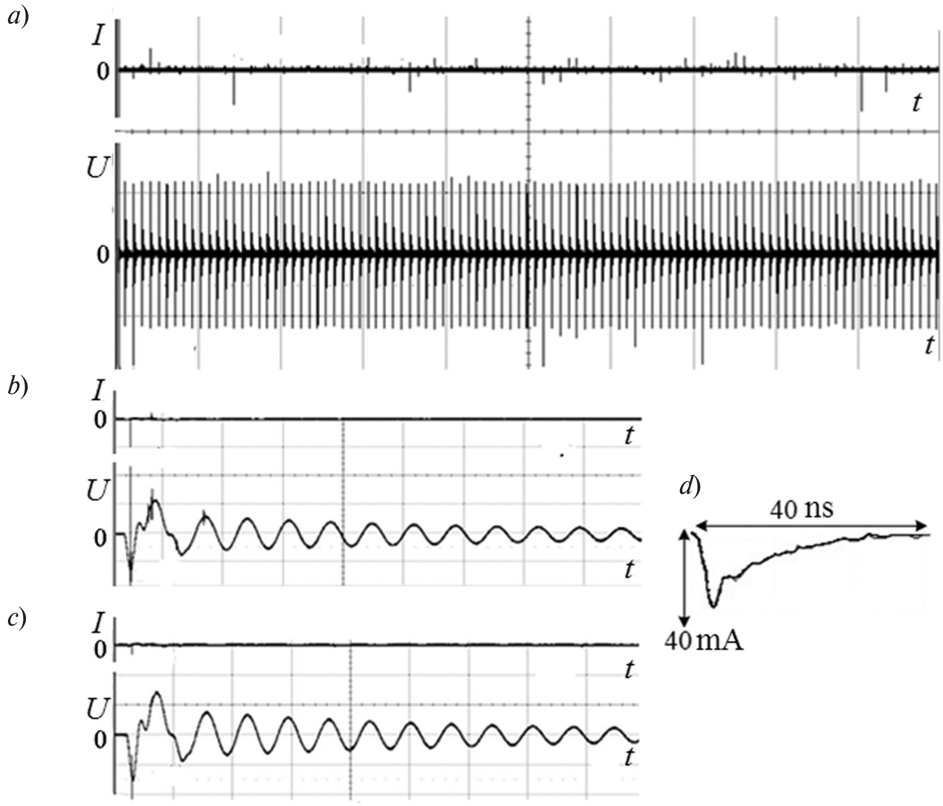

Рис. 3. Current and voltage waveforms of a negative asymmetrical barrier discharge evolving in the small spherical electrode, with different sweep speeds $\Delta t/n$. The waveforms were recorded for a sequence of pulse trains (*a*), and for a single pulse train: with a pair of pulses (*b*) and with a single current pulse (*c*); the shape of a single discharge current pulse ($U_1 \approx 11.8$ kV) (*d*) is also shown;
$\Delta t/n$ = 10 ms/div (*a*), 10 μs/div (*b*, *c*)

mum, and the voltage continues to increase after this breakdown (Fig. 3, *c*). This is because the amplitudes of the overvoltage pulses are small for the range considered.

With a positive pulse, the electric field of the surface charge is summed with the external field, and thus the resultant field strength in the gap reaches the breakdown value. This leads to the positive breakdown immediately after the negative in almost every case (see Fig. 3, *b*). Fig. 3, *a* shows that a positive pulse is observed for all relatively large negative current pulses, even if it is substantially smaller. There is no positive breakdown immediately after the negative only in very rare cases (see Fig. 3, *c*). This is most likely due to the statistical nature of initiation of avalanches by free electrons.

It can be seen from the waveforms in Fig. 3, *b* that an avalanche developing results in a spike in the voltage waveform. The short duration of the spikes indicates that they are actually caused by avalanches, and do not represent random electromagnetic interference. A spike can have the same and the opposite sign as the current pulse. No correlation was observed between the amplitude of the current pulse and the width of the spike, and in some cases, spikes were also observed in the absence of current pulses (for example, in the second positive half-cycle of the waveform, see Fig. 3, *b*). In this regard, such spikes can be considered to be indicators of low-intensity avalanches. Avalanche noise is the apparent reason for a small fluctuation in the voltage amplitude of the first pulse of the train, observed in the waveform in Fig. 3, *a*.

Fig. 4 shows the electrical characteristics of a positive asymmetrical barrier discharge, which is approximately the same as the negative one (see

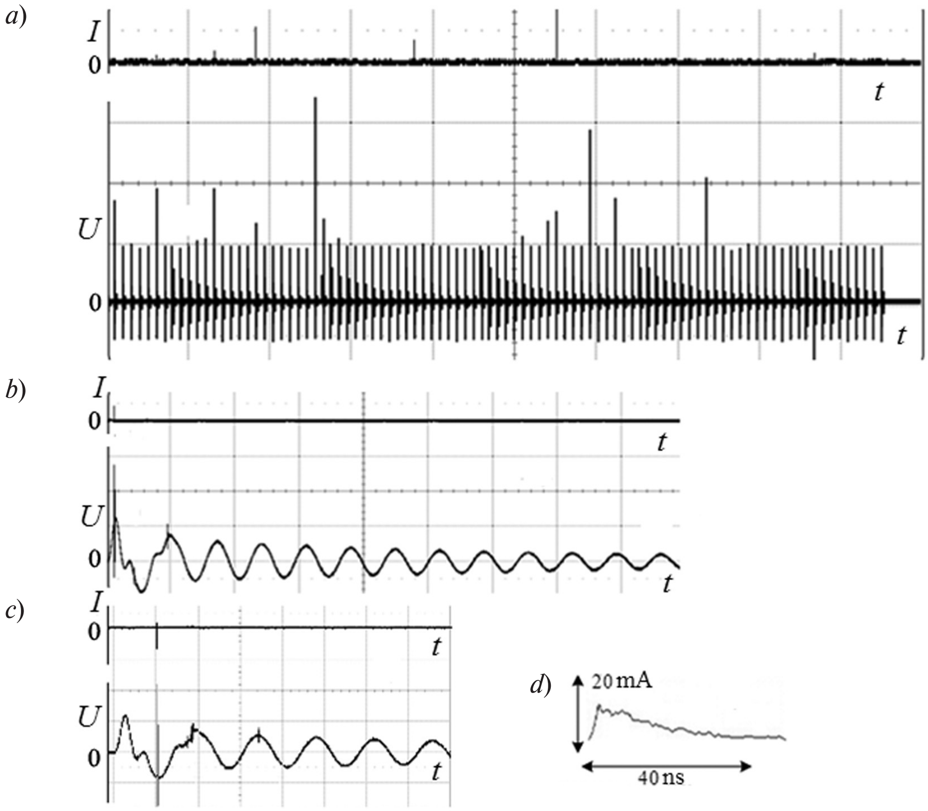

Fig. 4. Current and voltage waveforms with a positive asymmetrical barrier discharge evolving, for different sweep speeds $\Delta t/n$. The waveforms were recorded for a sequence of pulse trains (*a*) and for a single pulse train (*b*, *c*); the shape of a single discharge current pulse ($U_1 \approx 13.2$ kV) (*d*) is also shown;
$\Delta t/n = 10$ ms/div (*a*), 10 μs/div (*b*), 5 μs/div (*c*)

Fig. 3), but with an external voltage exceeded by 0.5 kV. It can be seen from the figure that for a discharge excited by positive pulses, the discharge current is a sequence consisting almost entirely of positive pulses, similar in their parameters to the current pulses for a negative discharge. Pulses of negative polarity could be "caught" in the millisecond range very rarely (once per dozen waveforms), and only in the second, negative pulse of a train (Fig. 4, *c*). The waveform in Fig. 4, *a* shows that current pulses exist only for some high-voltage trains of the sequence, even though voltage spikes (without a current pulse) can also be observed in subsequent negative half-periods (see Fig. 4). Very large spikes are also present on the voltage waveform with no current spikes at all, which was not observed for the negative discharge.

Thus, there is a significant difference between negative and positive asymmetrical barrier discharges. It should be emphasized here that, generally speaking, breakdowns in the same direction, observed for a positive discharge, contradict the very principle of the barrier discharge: current flow in the presence of dielectric layers on the electrodes is ultimately due to charges accumulating on these layers, and a reverse current phase is necessary in continuous mode to neutralize these charges and to accumulate charges of the opposite sign [8, 10].

We could hypothesize that such a difference is due to the difference in the relaxation processes of surface and space charges in both cases, but even rough estimates indicate that these processes should be close. Indeed, with a positive barrier discharge evolving, a "positive" avalanche, developed from the first pulse of a train while moving from the flat electrode to the spherical one and depositing its negative electron charge on the sphere, should leave the same distributed positive ion charge (the avalanche's "ion sheath") in the interelectrode gap.

During the remainder of the first pulse ($\approx$ 2 μs, see Fig. 4, *b*), positive ions are moved by the field in the direction opposite from the sphere. Assuming (for a rough estimate) that the space- and time-averaged field strength at the surface is about 20 kV/cm, and the mobility of positive ions is 1.25 $cm^2/(V \cdot s)$ [11], we obtain a drift velocity value of approximately $2.5 \cdot 10^4$ cm/s and a sufficiently large ($\approx$0.05 cm) distance of the front of the sheath (with the highest ion concentration) from the surface of the sphere. Accordingly, the "tail" of the ion sheath falls on the surface of the flat electrode, if the avalanche was initiated near it, and charges the electrode positively. However (see Fig. 4, *b*), the second, negative pulse (that has almost the same amplitude as the positive one) lasts two times longer, so when the ions move back to the surface of the sphere during this pulse, the corresponding distance must be substantially larger, equal to $\approx$ 0.2 cm. Thus, by that time, the majority of the positive space charge of ions should fall on the surface of the sphere and neutralize the part of the electron charge that was deposited there.

Subsequently, the positive and the negative pulses of the train last the same time, and the remaining small ion cloud makes damped oscillations, spreading due to diffusion, primarily in the radial direction. For the above ion mobility value at T = 300 K, the diffusion coefficient D = 0.032 cm^2/s, and the characteristic spreading time (over which the cloud size increases by $\sqrt{2}$) with an initial cloud radius of 50 μm is $t \approx$ 20 μs, i.e., of the order of the train duration. The spread then grows as $\sqrt{t}$, and the size $\approx$ 0.1 cm will be reached in a time $t \approx$ 20 ms. It is likely that a small number of ions neutralizing the negative surface charge will be added to the spherical electrode during this process of diffused spreading of the initial positive avalanche.

Thus, the charge of a single avalanche is mostly but not completely (this is of the essence) neutralized on the electrodes and in the interelectrode gap during the period between the trains. So subsequent avalanches of the same polarity increase the charge, until the charge that has increased after the latest avalanche increases the field of the negative pulse so that a breakdown with a "negative" avalanche (moving towardss the plate and carrying a negative electron charge) occurs, with a current pulse of negative polarity (see Fig. 4, *c*) neutralizing the accumulated charges. The fact that such breakdowns were extremely rare in the given conditions gives us grounds to conclude that, due to a good combination of the amplitude and the form of the train pulses (short first pulse), the neutralization of charges occurs quite effectively on its own.

The picture observed for a negative barrier discharge, when a negative avalanche formed during the first negative pulse of the train moves towards the plate and transfers its electronic charge to it, and the avalanche's ion sheath shifts towardss the spherical electrode, is only slightly more complex. In this case, as noted above, a "positive" avalanche moving towards the spherical electrode is formed, usually in the middle of the second, positive pulse (while the surface charge field is still large). After the avalanche has transferred its electron charge to the electrode, its ionic sheath, together with the remainder of the sheath from the first avalanche, moves towardss the plate under the action of the field during the second half of the positive pulse ($\approx$ 4 μs), and then to the spherical electrode again during the third pulse ($\approx$ 8 μs). As a result of the movement of the second sheath, the majority of its ions must reach the spherical electrode and neutralize the main part of the negative charge of the second avalanche during the third pulse.

A more complex picture should be observed in those modes of negative barrier discharge when the avalanches moving towards the flat electrode do not reach it because ionization stops in a weak field in this part of the discharge gap. In this case, the avalanche current is generated by the drift motion of the electrons that have already formed towards the plate. But since the probability that electrons attach to oxygen and nitrogen in air under atmospheric pressure is high (the attachment rate is $2 \cdot 10^8$ s^{-1} [8, 12]), they quickly turn into negative ions. In general, the presence of a space charge of slow negative ions considerably complicates the picture of charge relaxation in all cases when such ions are formed.

However, since the glow from the avalanches is typically adjacent to the surface of the flat electrode in both positive and negative asymmetrical barrier discharges for the modes under consideration, the neutralization efficiencies of the charges should not differ greatly. It is more likely that the reason for the observed difference is due to their asymmetry. In a negative discharge, the electron charge is deposited on the plate, where the field of the applied voltage pulse is minimal, and the additional field generated by the charge, which is summed with the field of the second, negative pulse, is the factor determining avalanche breakdown during this pulse. Conversely, the avalanche in a positive discharge carries an electron charge to the spherical electrode, where the field strength of the applied pulse is maximal and the addition from the surface charge has little effect on the formation of avalanches.

Conclusion

The findings we have obtained on asymmetrical barrier discharges helped establish a number of important regularities in discharge evolution, indicating that a stable homogeneous (avalanche) form of these discharges, which has significant advantages for practical applications, can be generated. Avalanche discharge likely occurs with an appropriately selected type of voltage supply, which ensures effective mutual compensation of volume and surface charges, especially for a positive discharge.

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THE AUTHORS

KORENYUGIN Dmitriy G.
Peter the Great St. Petersburg Polytechnic University
29 Politechnicheskaya St., St. Petersburg, 195251, Russian Federation
polarisdk@mail.ru

MARTSINOVSKY Artemiy M.
The Ioffe Institute of the Russian Academy of Sciences
26 Polytekhnicheskaya St., St. Petersburg, 194021, Russian Federation
amartsinovsky@gmail.com