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Ionization processes in spark discharge plasmas

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Abstract. The dominant mechanisms of electron generation and loss during different phases of spark discharge are considered. The focus is on spark formation in long non-uniform-field air gaps through leader and streamer processes. The streamer development in dry air and the effects of humidity, temperature and density on the properties of a long positive streamer in air are discussed. The ionization kinetics in the leader channel in long air gaps is described. The ionization mechanisms which lead to the re-breakdown within a post-arc air channel and to the streamer breakdown in undisturbed air and in Ar are presented.

1. Introduction

Experimental study of spark discharges in gaseous gaps (for the most part in air gaps) has a long history [1–4] in connection with the practical problems of dielectric behaviour of external insulation. However, properties of the spark discharge plasmas are poorly understood. It is hard to carry out any experiment with long sparks since high temporal resolution (down to 10^{-9} s) and spatial resolution (down to 10^{-2}–10^{-3} cm) are required; it is difficult to predict the spark path and to observe the spark during its development because of the low intensity of the light emitted by the spark channel during the first phases of its development. In addition, it is difficult to use sensitive equipment when high impulse voltages (up to 10^6 V and over) are applied to generate long sparks under laboratory conditions.

Thus, it is no wonder that a great deal of effort has been devoted to computer simulation of the development of the spark discharge (primarily the streamer development) which can provide insight into the fundamental processes on a microscopic scale during different phases of the spark formation. The simulation of long ($\gg$1 cm) sparks is a particularly severe problem because the spark development in long gas gaps is governed by a complex interaction of electromagnetic, hydrodynamic and kinetic elementary processes. Therefore, different phases of a spark in long non-uniform air gaps are usually simulated by simplifying a kinetic model for electron– and ion–molecule processes [3, 5, 6]; for instance, the plasma in the spark channel is assumed to be composed of electrons and one-species positive and negative ions.

Recently a numerical calculation which was based on a detailed kinetic model was used to simulate some phases of the spark development in long air gaps [7–12]. It turned out that the conventional models of spark discharge contain a number of misunderstandings concerning the dominant mechanisms of electron generation and loss which govern the plasma conductivity and strongly affect the observed phenomena.

The aim of this paper is to give the present view of the processes which control electron density and plasma conductivity in the curse of spark formation. The focus is on spark discharge in long non-uniform-field air gaps. In section 2, the fundamental concepts of pre-breakdown phenomena and spark breakdown in long gas gaps are given. The dominant mechanisms of electron generation and loss during the streamer development in dry air, humid air and heated (up to 1000 K) air are discussed in section 3. A kinetic model for the ionization processes in the leader channel in long air gaps is considered in section 4. The microscopic mechanisms of a streamer breakdown within a post-arc air channel, in undisturbed air and Ar are presented in section 5.

2. Fundamental concepts

Determining the conditions of a spark breakdown is a vital issue in the design of high voltage apparatus. In high voltage engineering, breakdown is understood as the formation of a highly conductive channel capable of carrying such a strong current that the voltage in the insulation gap sharply drops to produce short circuiting. For short circuiting to occur, the resistance of a channel that has bridged the gap must become lower than that of the external circuit. The channel must be unstable, and the rising current must reduce the voltage necessary to maintain the gas ionization; that is, an ionized channel must possess a falling current–voltage characteristic.

Spark discharges are generally studied in long ($>$1 cm) gaseous gaps at atmospheric pressure and above it. It is easier to initiate a spark in a gap with a strongly non-uniform distribution of the electric field between the electrodes, because this requires a lower voltage. Lightning is believed to be the longest (tens of kilometers) spark discharge in nature. As for their appearance, a laboratory spark and lightning have much in common, only differing in the scale.

In a non-uniform-electric-field gap, only in the vicinity of the small radius electrode is the external field sufficiently strong in order to ionize the gas. Therefore, the plasma channel can bridge the gap only due to the propagation of an ionizing wave which should be generated in the region of strong external field in the vicinity of the small radius electrode. Owing to high conductivity of the plasma channel, the initial potential can attach itself to the ionizing front, thus enhancing the local electric field. This is why an ionizing wave is able to propagate through a weak external field in the gap.

A classic example of the propagation of an ionizing wave in a gaseous gap is the streamer process. The foundation for a streamer theory was laid by the work of Loeb, Meek and Raether [1–3] in the 1940s. Later investigations provided much experimental and theoretical evidence that considerably changed some estimates and even fundamental concepts, but the basic ideology of this theory remained as before [4].

A streamer is a narrow plasma channel which can develop from an electrode of large curvature and propagate in low electric fields by reproducing high field at the streamer head. Ionization processes (electron impact ionization of neutral particles) take place only in this region, whereas the plasma in the streamer channel is in a decay regime. The reduced electric field E/N (N is the gas number density) is believed to range up to 10^3 Td (1 townsend (Td) is 10^{-17} V cm^2) at the streamer head and to be around 20 Td in the channel. Streamers can be as great as 1–10 m in length and electron density in the streamer channel is assumed to lie in the range 10^{14}–10^{15} cm^{-3}. During the streamer propagation the electron temperature T_e in the streamer channel rises to around 1 eV whereas the gas temperature T does not change.

After bridging a gaseous gap the streamer channel can spontaneously decay or transform into an arc. The latter case is referred to as a streamer breakdown. For a streamer breakdown to be developed in air or other molecular gases the gas temperature in the streamer channel after bridging the gap must rise to a high magnitude (5000–6000 K for air). In addition, the streamer channel must be heated for a short period of time when the plasma has no time to decay through electron attachment or electron–ion recombination. In air under standard conditions, the lifetime of the plasma lies in the sub-microsecond range; therefore, a streamer breakdown in air is a unique phenomenon requiring special conditions. However, a streamer breakdown can be easily observed in atomic gases [4] in which the plasma decay is much slower than that in molecular gases.

Breakdown in air gaps tens of metres long develops for hundreds of microseconds, whereas a lightning channel forms for a period of time up to 10 ms. Over these periods of time, the plasma in the channel, being in a weak electric field, can be maintained only due to gas heating. This is not the case during the streamer propagation. The process of the formation and propagation of the high-temperature channel is referred to as a leader process. This was discovered in the 1930s by Schonland [13] during the observation of lightning discharge. Several years later, Allibone in the UK [14] and Stekolnikov in Russia [15] independently confirmed the leader nature of a long laboratory spark. The present view of the leader process was gained by Komelkov in 1947–1950 [16] who experimentally showed that the leader development is accompanied by repeated streamer outburst from the channel front end known as leader head. The streamer velocity is one to three orders of magnitude higher than the leader velocity. The total streamer current provides the leader channel with power, heats it and maintains the plasma conductivity at a sufficiently high level. At every instant the streamer zone of the leader usually contains a great number (up to 10^4–10^5 in air) of streamers.

3. Streamer process

There is a strong interplay between kinetic processes at the streamer head and those in the streamer channel. Electron-impact processes (ionization, excitation and dissociation of molecules) in a high electric field at the streamer head provide the channel with charged particles, excited molecules and atoms. The rate of the plasma decay at a low field in the channel depends both on the electron density n_e and on the densities of atoms and excited molecules which can cause electron detachment from negative ions. In turn, electron density in the streamer channel and the channel conductivity affect the electric potential at the streamer head and therefore the rate of electron-impact processes in this region.

3.1. The streamer in long air gaps under standard conditions

In long air gaps, electron-impact processes at the streamer head are sufficiently well studied; the processes of electron loss and generation in the channel are less well understood. It is usually believed that the loss of electrons in the streamer channel in air is dominated by three-body attachment [3, 5, 6]

$$\mathrm{e} + \mathrm{O_2} + \mathrm{M} \rightarrow \mathrm{O_2^-} + \mathrm{M} \qquad k = k(E/N, T, \mathrm{M}) \tag{1}$$

where $\mathrm{M} = \mathrm{O_2}$ in dry air and $\mathrm{M} = \mathrm{O_2}$ and $\mathrm{H_2O}$ in humid air. Sometimes dissociative recombination with simple molecular ions (O_2^+, N_2^+)

$$\mathrm{e} + \mathrm{AB^+} \rightarrow \mathrm{A} + \mathrm{B} \tag{2}$$

is also considered.

There is reason to think that such simple models of the kinetic processes in the channel can fail to explain a number of the observations. Therefore, it is of interest to simulate the properties of a long streamer in air by taking into account a detailed kinetic model at the cost of a decrease in the model dimensionality. This approach was applied to study a long streamer in air gaps using a 1.5D simulation under the assumption that the radius of the streamer channel is fixed [7, 17] or taking into account the ionization expansion of the streamer channel in an approximate way [7, 8].

Based on a detailed kinetic plasma model, the 1.5D calculation showed that under standard conditions (i) the positive-ion composition changes during the streamer development and (ii) the loss of electrons in the channel is initially governed by dissociative recombination (process (2)) with complex ions. Process (1) becomes important only when the streamer conductivity decreases by more than a

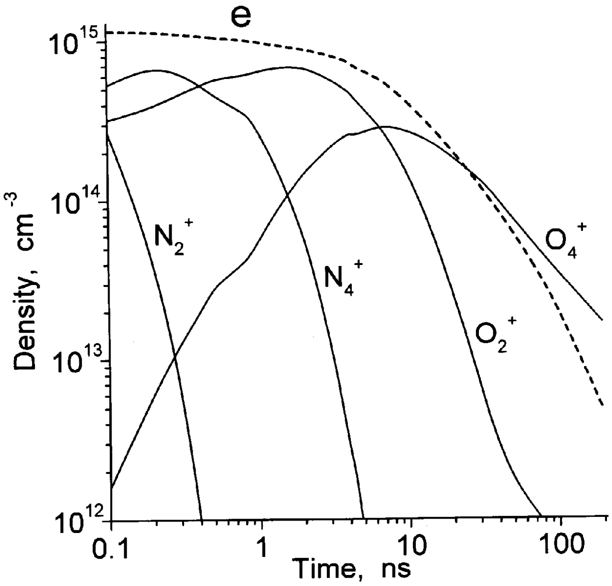

Figure 1. The evolution in time of densities of electrons, O_2^+, O_4^+, N_2^+ and N_4^+ in the positive-streamer plasma at a distance of 5.1 cm from the anode. The initial conditions used are a spherical anode of radius $R_a = 1$ cm, the applied voltage $U = 100$ kV and the channel radius $r_c = 0.03$ cm.

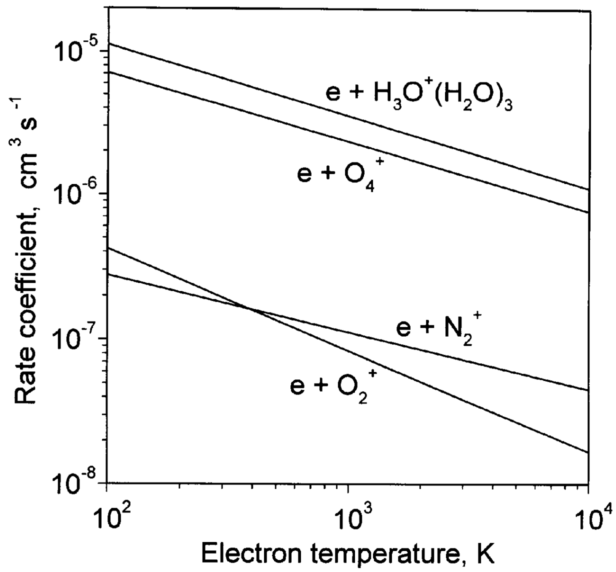

Figure 2. The rate coefficients for electron recombination with O_2^+ [18], N_2^+ [18], O_4^+ [18] and $H_3O^+(H_2O)_3$ [19] versus electron temperature.

factor of ten. Figure 1 shows the time evolution of densities of electrons and positive ions in the streamer plasma [7]. The dominant species of positive ions in the streamer channel in dry air are changed in time according to the following schema

$$N_2^+ \rightarrow N_4^+ \rightarrow O_2^+ \rightarrow O_4^+.$$

The N_2^+ ion produced by electron impact ionization at the streamer head rapidly reacts with N_2 in the three-body reaction

$$N_2^+ + N_2 + M \rightarrow N_4^+ + M$$

$$k = 5 \times 10^{-29} \times [300/T \text{ (K)}]^2 \text{ cm}^6 \text{ s}^{-1} \qquad (3)$$

then the O_2^+ ion is formed in the charge exchange reaction

$$N_4^+ + O_2 \rightarrow 2N_2 + O_2^+ \qquad k = 2.5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1} \quad (4)$$

and it is removed in the three-body reaction

$$O_2^+ + O_2 + M \rightarrow O_4^+ + M$$

$$k = 2.4 \times 10^{-30} \times [300/T \text{ (K)}]^3 \text{ cm}^6 \text{ s}^{-1}. \qquad (5)$$

Ion–molecule reactions changing the ionic species result in a marked variation in the average rate of the electron–ion recombination. The characteristic time of the ion–molecule reactions and that of electron loss in the streamer channel are of the same order. Therefore, there is reason to think that simplifying the positive-ion kinetics can result in large errors in the calculation not only of the composition of streamer plasma but also of the common streamer properties, namely the velocity of propagation, the conductivity and the limiting streamer length.

Under these conditions the electron density initially decreases by a factor of ten through the dissociative recombination with O_4^+ ions

$$e + O_4^+ \rightarrow 2O_2$$

$$k = 4.2 \times 10^{-6} \times [300/T_e \text{ (K)}]^{0.48} \text{ cm}^3 \text{ s}^{-1} \qquad (6)$$

with a rate which is approximately ten times that for simple ions (N_2^+, O_2^+) [18, 19] (see figure 2). Process (1) becomes important only about 100 ns later when the electron density falls by a factor of tens. Thus, the considered simulation shows that process (6) rather than (1) dominates during the first and more important phase of the plasma decay when the discharge energy supply changes drastically.

In long non-uniform air gaps under standard conditions, the simulation [7, 8] including ion–molecule reactions with cluster ions and ionization expansion of the streamer channel agrees well with the available experimental values of the total anode current, the streamer length and the average electric field in the streamer channel. For instance, the simulation of the streamer started from a spherical anode of radius $R_a = 12.5$ cm with the initial electric field of 40 kV cm^{-1} near the anode gave the value of the amplitude of the total anode current which differs by only about 10% from the measured magnitude (10 A [2, 20]). The calculated magnitude of the streamer length was 70 cm, which is also in good agreement with the result of the measurement [21] yielding 65 cm. The calculated average magnitude of the electric field in the streamer channel was about 5.3–5.4 kV cm^{-1} for a streamer of greater than 50 cm in length. This agrees well with numerous measurements of the average electric field in a streamer channel after bridging long air gaps at atmospheric pressure, which yield about 5 kV cm^{-1} [1, 2, 4, 22]. It should be noted that the agreement between the theory and the experiments for the above mentioned parameters cannot be obtained if the processes of cluster ions are not considered.

3.2. The effect of humidity

It has been found in experiments that the humidity retards the development of a streamer in air [1–3]. This reduces its limiting length and the total charge injected into a gap through

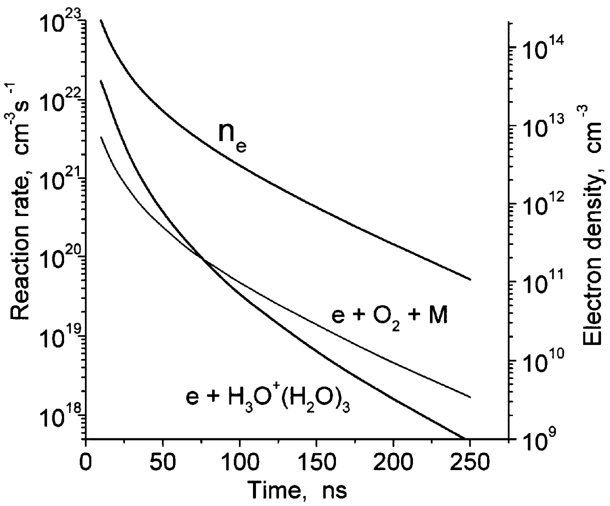

Figure 3. The evolution in time of n_e and rates of electron-loss processes in the streamer channel in humid air at a distance of 5.7 cm from the anode. The curves correspond to $R_a = 1$ cm, $U = 115$ kV, $p_w = 15$ mbar and $r_c = 0.03$ cm.

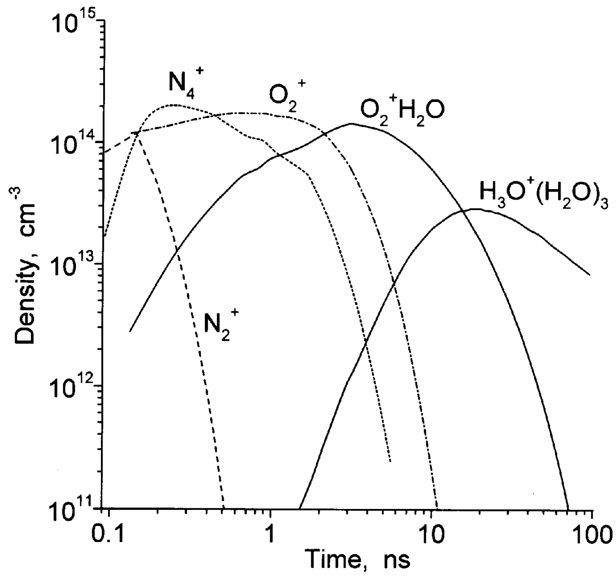

Figure 4. The evolution in time of densities of the dominant species of positive ions in the streamer channel in humid air at $p_w = 30$ mbar. The curves correspond to $R_a = 1$ cm, $U = 125$ kV and $r_c = 0.1$ cm.

the streamer channels. In this case the average electric field required for bridging the gap increases.

It is generally accepted [3, 23] that the main mechanism responsible for this effect is an increase in the rate of process (1) in humid air which is assumed to dominate the loss of electrons in the streamer channel. The rate of three-body electron attachment to O_2 for $M = H_2O$ is about ten times greater than for $M = O_2$ [24, 25]. It has been also proposed that the effect of humidity is due to decreasing rate of electron detachment from negative ions; a possible reason is clustering the negative ions [3] or quenching the metastable states of O_2 in collisions with H_2O

$$O_2(a^1\Delta_g) + H_2O \rightarrow O_2 + H_2O \qquad k = 5.6\times10^{-18}\ \mathrm{cm^3\,s^{-1}} \tag{7}$$

which play an important role in electron detachment from negative ions through the process [26]

$$O_2^- + O_2(a^1\Delta_g) \rightarrow e + 2O_2 \qquad k = 2\times10^{-10}\ \mathrm{cm^3\,s^{-1}} \tag{8}$$

The 1.5D simulation was used to explain the effect of humidity on the properties of a positive streamer in long air gaps [27]. Figure 3 shows the time evolution of the electron density and the rates of the dominant electron-loss processes in the streamer channel at a partial water vapour pressure $p_w = 15$ mbar. Figure 4 shows the evolution in time of the composition of positive ions under considered conditions. In humid air, the ion clustering reaction

$$O_2^+ + H_2O + M \rightarrow O_2^+ + H_2O + M$$

$$k = 2.6\times10^{-28}\ \mathrm{cm^6\,s^{-1}}\ (p \sim 1\ \mathrm{torr}) \tag{9}$$

is competitive to reaction (5) and the species of positive ions in the streamer channel change in time in the following way

$$N_2^+ \rightarrow N_4^+ \rightarrow O_2^+ \rightarrow O_2^+H_2O \rightarrow H_3O^+$$
$$\rightarrow H_3O^+H_2O \rightarrow H_3O^+(H_2O)_2 \rightarrow H_3O^+(H_2O)_3.$$

Conversion to a hydronium ion or its hydrate occurs via

$$O_2^+H_2O + H_2O \rightarrow H_3O^+ + OH + O_2$$

$$k = 3\times10^{-10}\ \mathrm{cm^3\,s^{-1}} \tag{10}$$

and

$$H_3O^+(H_2O)_n + H_2O + M \rightarrow H_3O^+(H_2O)_{n+1} + M$$

$$k = 3\times10^{-27}\ \mathrm{cm^6\,s^{-1}}(p \sim 1\ \mathrm{torr}). \tag{11}$$

Under these conditions the loss of electrons is mainly governed by the dissociative recombination with an $H_3O^+(H_2O)_3$ ion

$$e + H_3O^+(H_2O)_3 \rightarrow H + 4H_2O$$

$$k = 6.5\times10^{-6}\times[300/T_e\ (\mathrm{K})]^{0.5}\ \mathrm{cm^3\,s^{-1}}. \tag{12}$$

The rate of electron recombination with $H_3O^+(H_2O)_3$ ions is greater than that with O_4^+ ions [18, 19] (see figure 2). The acceleration of the plasma decay in the humid air decreases the conductivity of the streamer channel and as a consequence the electric field at the streamer head is reduced. Therefore, the averaged electric field required for bridging the gap increases with increasing humidity. The calculated humidity dependence of this electric field agrees qualitatively with the available observations [23].

The rate of three-body electron attachment to O_2 molecule also increases with humidity. However, this process becomes important only when the density of electrons in the channel falls by a factor of ten, as shown in figure 3. Similarly, electron detachment from negative ions can be neglected in humid air because of a low density of negative ions in the streamer plasma.

The considered effect becomes even more pronounced for streamers in flue gases which contain a great percentage (10–20%) of H_2O. This is important for impulsed positive corona discharges which are actively studied in connection with their use for flue gas cleaning from toxic components (NO_x and SO_2) [28].

3.3. Temperature and density effects

A study of the effects of temperature and density on the properties of a long streamer in air, besides its practical significance, provides insight into the mechanism of the streamer-to-leader transition. The mechanism of the transition from a cold streamer to the hot leader is still under discussion. It is clear that the plasma channel is finally heated to temperatures high enough for fast thermal ionization even in a weak electric field. A basic problem is to understand why during gas heating the streamer plasma does not disappear for a period of electron attachment and recombination in air under standard conditions.

Today it is generally accepted that the main mechanism responsible for maintaining the conductivity of streamers and starting the formation of a leader in air consists of thermal electron detachment from negative ions [3, 5, 6, 29]. According to this hypothesis the electron detachment compensates the losses of electron attachment and the streamer channel is conductive for a period of 1–10 μs, provided that the gas temperature within the channel is raised above a critical value of 1000–2000 K.

It was observed experimentally [30] that the conductivity of the positive-streamer channel in atmospheric-pressure air increases by a factor of 10^5 when the gas temperature grows from 290 K to 900 K. The density effect at room temperature was less pronounced than the total temperature effect was. The observed phenomena were explained by accelerating electron detachment from negative ions with increasing gas temperature.

In order to reveal the general discharge mechanism it is important to identify separately the density effect and the specific temperature effect, both of which contribute to the total temperature effect on the streamer properties at constant gas pressure. The specific temperature effect can be due to the temperature-dependence of the rates of collisional processes, such as electron detachment; the density effect can result from a difference between the density-dependence for the rates of two-body processes and that for the rates of three-body processes.

A 1.5D simulation [9] based on a detailed charge-particle kinetic model was used to study the temperature and density effects on the properties of a long positive streamer under conditions of the experiment [30]. The results of this calculation qualitatively confirmed with the measurements of electric fields in the bridging gap and oscillograms of the charge transferred through the streamer to the cathode at different gas temperatures and densities. However, the explanation of the temperature effect which followed from the simulation differs from that which was suggested previously.

Figure 5 shows the temporal evolution of electron density and rate for each specific process of electron loss/generation in the streamer plasma. As mentioned above, under standard conditions, the electron density initially decreases by a factor of hundred through process (6). Process (1) becomes important only about 100 ns later.

A decrease in the gas density at room temperature decelerates three-body reactions with respect to two-body reactions. This slows down the plasma decay through electron attachment (process (1)) and, what is more important, changes the positive-ion composition in the

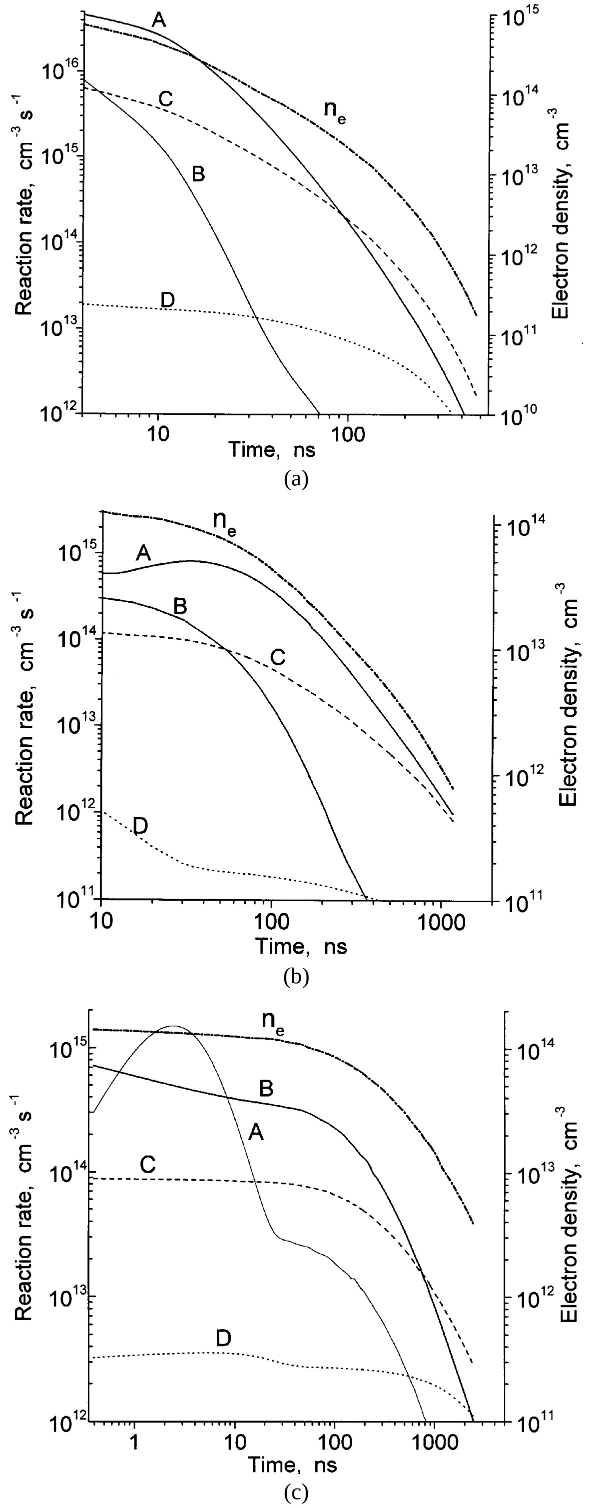

Figure 5. The evolution in time of n_e and frequency of each electron loss/generation processes in the streamer channel in dry air: (a) standard conditions, $p = 1$ bar and $T = 300$ K; (b) reduced density, $p = 0.33$ bar and $T = 300$ K and (c) reduced density, $p = 1$ bar and $T = 900$ K. The processes are: curve A, recombination with cluster ions (O_4^+ and N_4^+); curve B, recombination with simple ions (O_2^+ and N_2^+); curve C, three-body attachment to O_2; and curve D, electron detachment from O_2^-. The curves correspond to a 20 cm sphere-plane gap with $R_a = 1$ cm, $r_c = 0.03$ cm and $U = 104$ kV under standard conditions. At reduced gas density the voltage was changed to retain constant the ratio E/N.

channel plasma; the cluster ion (O_4^+) density decreases and the simple ion (O_2^+) one increases because of the deceleration of the three-body conversion processes (reactions (3) and (5)). Therefore, the apparent rate of electron–ion recombination also decreases with decreasing gas density. The deceleration in the plasma decay at lowered air density increases the conductivity of the streamer channel.

Gas heating up to 900 K results in the total decomposition of cluster ions through the processes which are reverse to reactions (3) and (5) [31, 32]; this occurs because of the low dissociation energy of these ions (0.5 eV for O_4^+). Therefore, at higher air temperature and at atmospheric pressure both the electron–ion recombination and the three-body electron attachment to O_2 molecules slow down. The first process decelerates owing to the change in the positive-ion composition and the second one because of a decrease in the gas density. As a result, the streamer approaching the cathode remains highly conductive. Figure 5(c) shows that electron attachment becomes important for $t > 1$ μs only. Although the rate of electron detachment increases drastically with increasing T in comparison with that observed at lower pressure (see figure 5(b)), this process is not essential.

To clarify directly the role of electron detachment in high-temperature air a simulation of the streamer properties was performed at 900 K in which this process was not taken into account. The obtained results have shown that neglecting electron detachment does not affect the main manifestations of the temperature effect on the streamer properties; that is, the observed temperature effect is primarily explained by deceleration of electron–ion recombination because of the transformation of positive cluster ions into simple ions.

4. Leader process

Leader propagation is an important phase in the discharge breakdown of very long (>1 m) air gaps. The leader channel is characterized by a high gas temperature (1000 K at the leader tip and 5000–6000 K in the long-lived regions of the leader) which causes a rise in the lifetime of the leader plasma by many orders of magnitude. In order to maintain the plasma during the leader development (for 10^{-3}–10^{-2} s), it is necessary not only to slow down the loss of electrons but also to ionize neutral particles.

The current density j in the leader channel is governed not by the plasma conductivity but rather by the ionization processes in the streamer zone in front of the leader tip. The electric field in the channel varies self-consistently with n_e and is determined by the value of j. As a result, the processes of electron generation and loss in the channel largely control the voltage drop along the long leader channel and the spark-over voltage of long air gaps.

The known models [1, 3, 5, 6, 33] of the leader channel in long air gaps generally assume that electron generation is dominated by electron-impact ionization and that the ratio E/N remains constant (50–80 Td) during the development of the leader. The main action of gas heating on the leader plasma properties is assumed to be due to electron detachment from negative ions and owing to a decrease in gas number density N at constant pressure. It is assumed that other ionization processes and the influence of the gas temperature on the ionization kinetics due to a change in the gas composition are neglected. However, it is well known that the last statement is not valid for high-temperature air in the local thermodynamic equilibrium (LTE). Nevertheless, this would be true if the time interval needed for response of gas temperature changes in the ionization kinetics is longer than the time of the leader development.

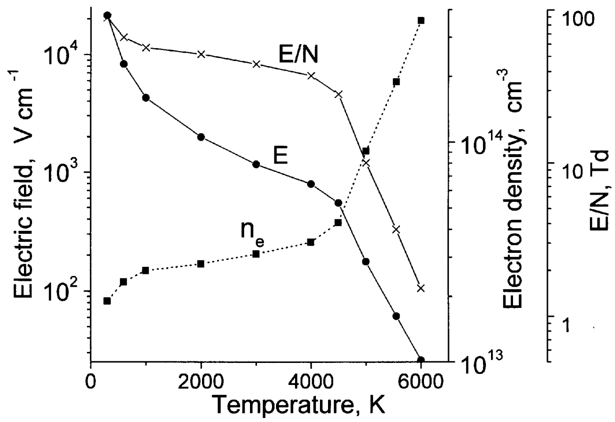

Figure 6. The quasi-steady-state values of E, E/N and n_e in the leader channel versus T. The curves correspond to a leader current of 1 A and to a thermal channel radius of 0.1 cm in a long air gap.

A detailed kinetic model for the ionization processes in high-temperature air in an electric field was used to estimate the response time for the leader plasma and by this means to identify the dominant mechanisms of electron generation and loss in the leader channel in long air gaps [10]. Numerical simulation has shown that the typical relaxation time (20–30 μs) of the electrical parameters in the leader channel is much less (by a factor of ten or more) than the development time of the leader in the 5–10 m gaps at the minimal breakdown voltage. Therefore, in the leader channel the density of electrons, the conductivity and the electric field should nearly follow the local value of the gas temperature. The calculation also showed that the mechanism of ionization processes in the channel is changed drastically with increasing gas temperature and subsequently affects the leader properties.

Figure 6 shows the calculated quasi-steady-state values of the electron density n_e and the electric field in the leader channel versus the gas temperature T. It is evident that the hypothesis that the value of E/N remains constant during the leader development is not true because of the effect of the gas temperature on the ionization kinetics. In the range 1000–4000 K the value of E/N falls from 55 to 37 Td because the rate of electron-impact ionization increases with increasing T due to the formation of NO molecules having low ionization energy (9.26 eV). Thus, the ionization of NO becomes a dominant mechanism of electron generation in the channel. At $T > 4500$ K the dominant ionization mechanism becomes the associative ionization

$$O + N \rightarrow e + NO^+ \qquad (13)$$

which leads to a sharp drop in E/N (to 1.5 Td at 6000 K). The rate of this process is independent of the field because the densities of O and N depend only on T. In this case, the field in the leader channel governs the electron density only through the T_e-dependent rate of the electron loss which is dominated by electron recombination with NO^+ ions.

The gas temperature in long-lived sections of the leader channel exceeds 5000 K even at $t > 200$–$300\ \mu$s [1–3] when the leader is a few metres in length. In very long leaders with a life of 10 ms, temperatures >5000 K are typical for the channel overall except for the transient region 1–3 m in length behind the leader tip. This allows the leader to bridge air gaps hundreds of metres in length at an average electric field of the order of 10^2 V cm^{-1}, which was observed experimentally [34, 35]. It should be noted that these experiments cannot be explained using the hypothesis that the value of E/N remains constant in the leader channel.

5. Streamer breakdown

After bridging the gaseous gap the streamer channel can spontaneously decay or transform into arc which is able to maintain the current even at a drastic decrease in the applied voltage. The latter case is referred to as a streamer breakdown which is the fastest way to form the spark channel in long gaseous gaps. Let us consider the conditions when a streamer breakdown can develop and ionization processes which are the basis of this phenomenon.

5.1. Re-breakdown within a post-arc channel in long non-uniform air gaps

Prior heating of air favours producing a spark breakdown. For instance, this occurs during the re-breakdown of air gaps after arc extinction which is encountered in circuit breakers, power transmission lines and in the channel of many-component-lightning discharge. In these cases the re-breakdown develops within the post-arc channel which can differ from the ambient air by the gas temperature, density, chemical composition and degree of ionization. Therefore, the re-breakdown threshold in air gaps after an arc discharge can be many times smaller than the breakdown voltage in undisturbed air.

A conventional approach to study the re-breakdown in air gaps consists of modelling the decay of the gas temperature in the post-arc channel and calculation of the breakdown voltage $U_{br}(T)$ under the assumption that

$$U_{br}(T) = U_{br}(T_0)N/N_0 = U_{br}(T_0)T_0/T \qquad (14)$$

where T and N are the current values of the gas temperature and number density in the decaying channel at the moment when the high voltage is re-applied and T_0 and N_0 are those in the undisturbed air; that is, the re-breakdown reduced field E/N is constant. Assumption (14) implies that (i) the temperature effect reduces to the density effect and (ii) the re-breakdown mechanism, like the primary breakdown mechanism, is associated with the ordinary leader process.

A 1.5D simulation [11] studied the mechanism of the re-breakdown along the long post-arc channel in non-uniform-field air gaps and the fundamental ionization processes which govern this phenomenon. The dynamics of the channel cooling was not considered; instead, the initial values of T and of the electron density n_e and the initial gas composition

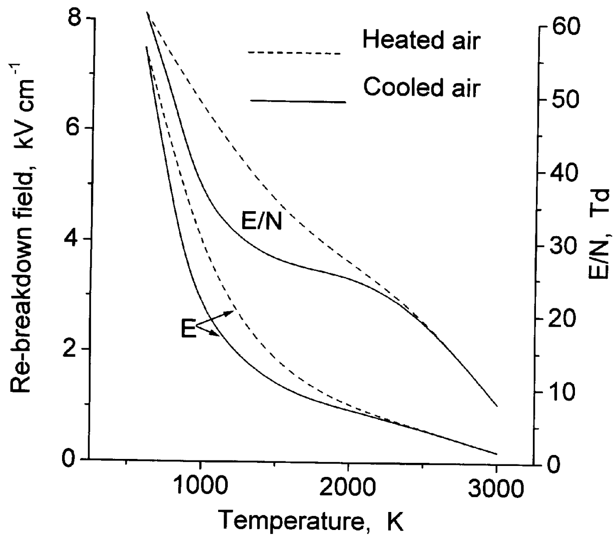

Figure 7. The breakdown values of E and E/N versus the initial gas temperature. Solid curves correspond to cooled air (re-breakdown in a post-arc channel) and dashed curves correspond to heated air.

were taken as input parameters. An auxiliary calculation showed that the initial densities of electrons, O and N can be assumed to be equal to those under the LTE conditions at the current temperature T. The same could be said for the density of NO only at $T > T_{max} = 3000$–3500 K. (At this temperature the LTE density of NO has a maximum.) At lower temperatures the loss of NO through chemical reactions was very slow; therefore, in this case the initial number of NO was assumed to be ‘frozen’.

The simulation [11] showed that in nature the re-breakdown in a post-arc channel in long non-uniform air gaps generally exhibits two distinct phases. The first phase consists of an ionizing wave traversing the gap along the post-arc channel. This wave is qualitatively similar to a streamer propagating in ambient air. However, the quantitative characteristics of this wave depend strongly on the initial values of T and n_e in the channel. The ionizing wave reaching the cathode induces a return wave. After bridging the gap by the return wave, the axial distributions of n_e and E in the channel become uniform. The second phase consists of a uniform increase in electron density and gas heating along the whole length of the channel which can be simulated by neglecting the longitudinal inhomogenity of the plasma.

Figure 7 shows the calculated re-breakdown threshold values of E and E/N as a function of $T(t = 0)$. In the range 600–3000 K the value of E falls from 7.5 to 0.2 kV cm^{-1}. An important point arising from this calculation is that the value of E/N does not remain constant but falls from 60 to 8 Td. This is due to changes in the ionization kinetics in high-temperature air.

At $T = 3000$ K the dominant ionization mechanism is process (13) and the calculated re-breakdown field is of the same order of magnitude as typical fields in an arc channel. Therefore, one should say that at $T > 3000$ K the recovery of the dielectric properties of air in a cooled post-arc channel does not yet occur.

Cooling of gas in the post-arc channel down to temperatures $T < 3000$ K inhibits process (13) and leads to a

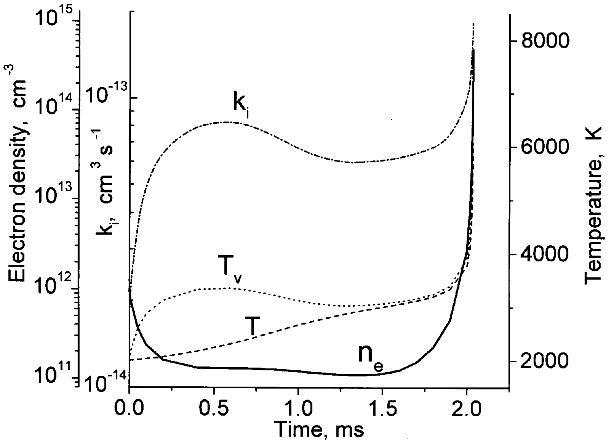

Figure 8. The evolution in time of n_e, k_i, T and T_v during the re-breakdown within a post-arc air channel at $T(t = 0) = 2000$ K and $E = 1$ kV cm^{-1}.

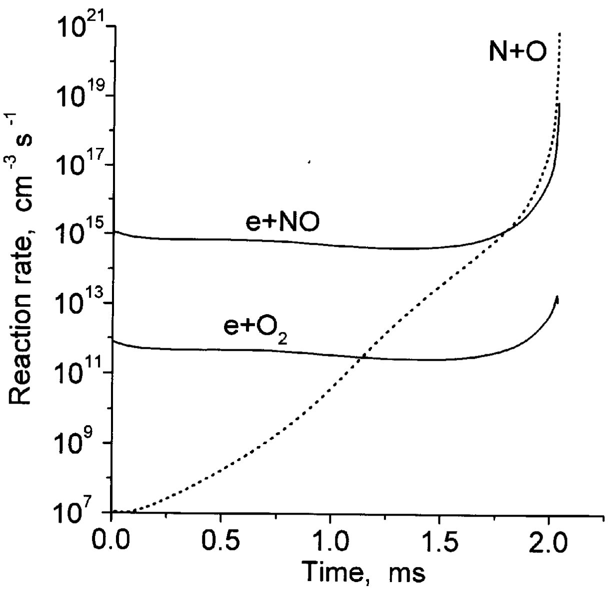

Figure 9. The evolution in time of the rates of each ionization process (e + NO $\rightarrow$ 2e + NO$^+$; e + O_2 $\rightarrow$ 2e + O_2^+ and N + O $\rightarrow$ e + NO$^+$) during the re-breakdown within a post-arc air channel. The curves correspond to the same conditions as those in figure 8.

sharp rise in the re-breakdown reduced electric field $(E/N)_{rb}$ which is required to maintain electron impact ionization of molecules. The calculations show that at $3000 > T > 1000$ K the dominant ionization mechanism is ionization of NO molecules having low ionization energy. At $T < 1000$ K there is another sharp rise in $(E/N)_{rb}$ because the rate of electron attachment to O_2 is no longer balanced by the rate of detachment from negative ions. At such large fields ionization is dominated by electron impact of O_2.

It is of interest that at $T < 3000$ K the initial density of NO in the post-arc channel is greater than that under the LTE conditions. This leads to a hysteresis effect as is shown in figure 7; that is, at a given gas temperature the breakdown voltage for cooled air with the enhanced density of weakly ionized NO molecules is lower than that for heated air when the density of NO corresponds to the LTE conditions.

In order to identify the dominant mechanism of the re-breakdown development the time dependent evolution of n_e, the ionization rate coefficient k_i, the gas temperature T and the vibrational temperature T_v of N_2 molecules are plotted in figure 8. Figure 9 shows the evolution in time of the rates of different ionization mechanisms in the channel under these conditions. It follows from this simulation that the rise in n_e at $t > 1.5$ ms is initially associated with an increase in T_v which causes an overpopulation of high-energy electrons and an increase in k_i due to second-kind collisions between electrons and vibrationally excited N_2 molecules. The electron density rises sharply when T exceeds 3000–3500 K. Here, the associative ionization (process (13)) becomes a dominant mechanism of ionization due to the thermal dissociation of O_2 and N_2 and because of a strong temperature dependence of the rate of this process. The ionization of neutral particles by field-heated electrons is no longer of significance; that is, the re-breakdown has been finished.

5.2. Streamer breakdown in long air gaps under standard conditions

The problem of streamer breakdown in long (>10 cm) air gaps at atmospheric pressure has not yet been solved. It is well known that, in a microsecond range, the breakdown of a long gap is governed by the leader process [1–4]. However, for nanosecond pulses, the breakdown cannot be attributed to the leader mechanism because the development of the leader takes a substantially longer time. Voltage pulses with a nanosecond front occur in the return stroke of the subsequent components of a lightning and are characteristic of some types of overvoltage pulse acting on the insulator gaps of high-voltage objects.

The streamer breakdown in long air gaps under standard conditions was studied by numerical modelling [12]. It was shown that this phenomenon is in some ways similar to the re-breakdown within a post-arc channel. A distinguishing feature of a streamer breakdown in ambient air is that the channel must be formed and heated for a short period of time when the plasma has no time to decay through electron attachment or electron–ion recombination. In air under standard conditions, the lifetime of plasma lies in the submicrosecond range. In order to bridge a gap for the considered periods of time one needs very strong external electric fields. The calculation [12] shows that the average breakdown field under standard conditions is around 25 kV cm^{-1} which is 5–6 times higher than that for the well studied leader breakdown.

The basis for the streamer breakdown in long air gaps is the following sequence of kinetic processes. After bridging the gap, the deceleration of the plasma decay is due to gas heating. At $T \approx 10^3$ K electron attachment is completely balanced by electron detachment, whereas the rate of dissociative electron–ion recombination decreases many times because of the decomposition of complex positive ions. Gas heating to 2000–3000 K is greatly favourable to electron impact ionization due to the formation of NO molecules with low ionization energy. Finally at $T = 4500$–5000 K associative ionization (process (13)) becomes the dominant ionization mechanism in the channel.

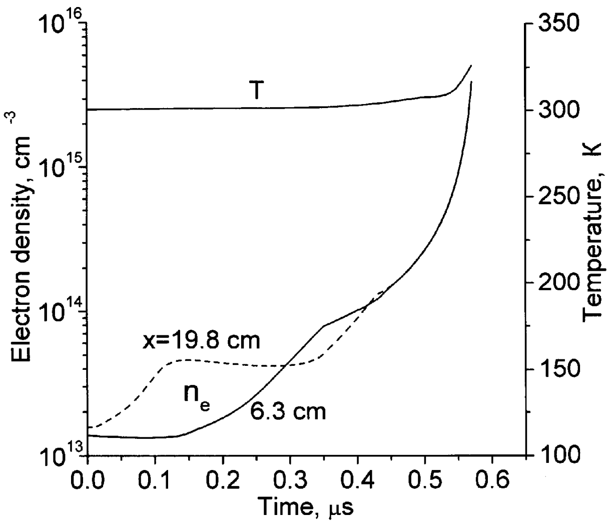

Figure 10. The evolution in time of n_e and T in the streamer channel in Ar after bridging the gap. The curves correspond to a 25 cm sphere–plane gap at $R_a = 0.5$ cm, $U = 22$ kV with a rise time of 1 μs and $r_c = 0.1$ cm.

The rate of this process is independent of the field because the densities of N and O depend only on T. The streamer breakdown is expected to be completed.

The dominant mechanism for a fast gas heating in the streamer-breakdown field is the conversion of electronically excited states of molecules which are populated by electron impact into heat via the quenching of excited molecules by neutral particles

$$A^* + B \rightarrow A + B. \quad (15)$$

The fraction of energy transformed from electronic states of molecules into heat through this process is poorly studied. Therefore, the accuracy of the results of the calculation [12] is limited. However, it seems that the main mechanisms of a streamer breakdown were revealed by this simulation.

5.3. Streamer breakdown in Ar

Bazelyan *et al* [36] observed a streamer breakdown in long non-uniform Ar gaps under standard conditions. A most surprising point was that the breakdown was produced in Ar when the discharge energy supply was too low to heat the gas in a typical streamer.

The 1.5D simulation [37] revealed a sharp distinction between the properties of long streamers in Ar and air. The rate of electron loss process in the streamer channel initiated in Ar is low. Moreover, there is a significant generation of electrons which is due to two-step electron-impact ionization through excited states of Ar

$$e + Ar \rightarrow e + Ar^* \quad (16)$$

$$e + Ar^* \rightarrow 2e + Ar^+. \quad (17)$$

It is important that the electron-impact excitation rates increase with increasing electron density; this is due to electron–electron collisions which drive the electron energy distribution towards a Maxwellian one. Figure 10 shows the calculated temporal evolution of n_e and T in the streamer channel after bridging the gap. It is evident that the breakdown is produced without any gas heating. This is due to an increase in the ionization rate with increasing electron density of the two-step mechanism of the electron generation and the effect of electron–electron collisions on the electron energy distribution. As a result, the calculated values of breakdown field and time are around 600 V cm^{-1} and 1 μs, in good agreement with the experiment in Ar [36] and in contradiction with the observations in molecular gases.

6. Conclusions

The properties of spark discharge plasmas are governed by a variety of the processes of electron generation and loss. The rates of these processes can depend on electric field, gas pressure and density, neutral-particle and ion composition, the vibrational and electronic excitation of molecules, the ionization degree etc. For instance, the propagation of a streamer in long gaseous gaps is caused by fast ionization at the streamer head and slow decay of the plasma in the streamer channel. A change in the positive-ion composition in the streamer channel results in the observed effects of humidity, gas temperature and density on the properties of streamers in long air gaps. The leader process and the spark breakdown in long air gaps are related directly with associative ionization in collision between O and N atoms which becomes the dominant ionization process only after gas heating up around 5000 K. During gas heating, formation of NO molecules through chemical reaction can accelerate ionization by electron impact. Non-equilibrium density of NO in post-arc channel can lead to the hysteresis for the breakdown threshold in high-temperature air. Fast gas heating due to quenching electronically excited molecules can favour the development of a streamer breakdown in long air gaps under standard conditions. Finally, two-step ionization in the streamer channel and the effect of Coulomb collisions can result in a streamer breakdown in long gaps in rare gases.

In summary, in order to simulate the development of spark discharge it is important to take into account a detailed model of electron– and ion–molecule reactions. Any simplification of the kinetic model should be verified.

Acknowledgments

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References

[1] Meek J M and Craggs J D 1978 *Electrical Breakdown of Gases* (New York: Wiley)

[2] Les Renardieres Group 1977 *Electra* **53** 31

[3] Gallimberti I 1979 *J. Physique* **40** C7 193

[4] Bazelyan E M and Raizer Yu P 1998 *Spark Discharge* (New York: Chemical Rubber Company)

[5] Bondiou A and Gallimberti I 1994 *J. Phys. D: Appl. Phys.* **27** 1252

[6] Goelian N, Lalande P, Bondiou-Clergerie A, Bacchiega G L, Gazzani A and Gallimberti I 1997 *J. Phys. D: Appl. Phys.* **30** 2441

[7] Aleksandrov N L and Bazelyan E M 1996 *J. Phys. D: Appl. Phys.* **29** 740

[8] Aleksandrov N L and Bazelyan E M 1996 *Plasma Phys. Rep.* **22** 417
[9] Aleksandrov N L and Bazelyan E M 1996 *J. Phys. D: Appl. Phys.* **29** 2873
[10] Aleksandrov N L, Bazelyan E M, Kochetov I V and Dyatko N A 1997 *J. Phys. D: Appl. Phys.* **30** 1616
[11] Aleksandrov N L and Bazelyan E M 1998 *J. Phys. D: Appl. Phys.* **31** 1343
[12] Aleksandrov N L, Bazelyan E M, Dyatko N A and Kochetov I V 1998 *Plasma Phys. Rep.* **24** 541
[13] Schonland B 1956 *Handbuch der Physik* vol 22 (Berlin: Springer)
[14] Allibone T and Schonland B 1934 *Nature* **134** 3393
[15] Stekolnikov I S, Beljakov A P and Mjakishev I N 1937 *Elektrichestvo* **8** 51
[16] Komelkov V S 1947 *Izv. Akad. Nauk SSSR Tekh.* **8** 955
[17] Aleksandrov N L, Bazelyan A E, Bazelyan E M and Kochetov I V 1995 *Plasma Phys. Rep.* **21** 57
[18] Mitchell J B A 1990 *Phys. Rep.* **186** 215
[19] Johnsen R 1993 *J. Chem. Phys.* **98** 5390
[20] Stekolnikov I S 1960 *The Nature of the Long Spark* (Moscow: Izdat Akademii Nauk SSSR) (Engl. transl. 1962 USA Department of Commerce FTD-TT-61-149)
[21] Brago E N and Stekolnikov I S 1958 *Izv. Akad. Nauk SSSR OTN* **11** 51
[22] Bazelyan E M and Razhanskii I M 1988 *Spark Discharge in Air* (Novosibirsk: Nauka)
[23] Phelps C T and Griffiths R F 1976 *J. Appl. Phys.* **47** 2929
[24] Smirnov B M 1982 *Negative Ions* (New York: McGraw-Hill)
[25] Aleksandrov N L 1988 *Sov. Phys.–Usp.* **31** 101
[26] Lowke J J 1992 *J. Phys. D: Appl. Phys.* **25** 202
[27] Aleksandrov N L, Bazelyan E M and Novitskii D A 1998 *Tech. Phys. Lett.* **24** 367
[28] Penetrante B M and Schultheis S E (eds) 1993 *Non-Thermal Plasma Techniques for Pollution Control (NATO ASI Series G: Ecological Sciences 34)* pts A, B (Berlin: Springer)
[29] Raizer Yu P 1991 *Gas Discharge Physics* (Berlin: Springer)
[30] Aleksandrov D S, Bazelyan E M and Bekzhanov B I 1984 *Izv. Akad. Nauk SSSR Energ. Transp.* N **2** 120
[31] Smirnov B M 1983 *Complex Ions* (Moscow: Nauka)
[32] Kossyi I A, Kostinsky A Yu, Matveyev A A and Silakov V P 1992 *Plasma Sources Sci. Technol.* **1** 207
[33] Ross J N, Jones B and Waters R T 1977 *Electra* **53** 85
[34] Anisimov E I, Bogdanov O V, Gayvaronskii A S, Kol'tzov V A and Ovsyannikov A G 1988 *Electrichestvo* **11** 55
[35] Gayvaronskii A S and Ovsyannikov A G 1992 *Proc. 9th Int. Conf. on Atmospheric Electricity (Sankt Petersburg, 1992)* (St Petersburg: Voeikov Main Geophysical Observatory) p 792
[36] Bazelyan E M, Goncharov V A and Gorunov A Yu 1985 *Izv. Akad. Nauk SSSR Energ. Transp.* N **2** 154
[37] Aleksandrov N L, Bazelyan E M and Kochetov I V 1998 *Abstr. 14th Eur. Sect. Conf. on At. Mol. Physics Ion. Gases (Dublin)* ed D Riley *et al* (Belfast: European Physical Society) p 170