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ELEMENTARY PROCESSES IN PREBREAKDOWN PHENOMENA IN THE ATMOSPHERE

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The effect of negative oxygen ion destruction upon breakdown conditions in atmospheric air is analyzed. It is shown that ozone accumulation due to plasmochemical reactions occurring in ionized air produces a reduction in the breakdown voltage, related to negative O^- ion destruction upon collision with ozone molecules under realistic conditions. A relationship is derived for electric field breakdown intensity and ozone molecule lifetime for the real atmosphere.

The simplest condition for electrical breakdown in the atmosphere, as in any other electronegative gas, requires equality of the electron collision molecule ionization rate to the rate of electron attachment to molecules. According to this condition the breakdown electrical field intensity for dry air under normal conditions is 25.5 kV/cm. However the phenomenon of large scale electrical breakdown in the real atmosphere, i.e., lightning, can occur at significantly lower electric field intensities. One cause of reduced electrical strength of the air atmosphere is related to the medium's chemical composition, involving plasmochemical processes under the action of an external electric field. This leads to a change in the balance between the processes of formation and neutralization of free electrons, which opens, in particular, channels for the destruction of negative ions, the presence of which encourages breakdown conditions. Below we will analyze in detail this mechanism for reduction in electrical strength of such molecules. Special attention will be given to analysis of available information on the characteristics of these elementary processes and selection of the most reliable data.

In analyzing this phenomenon we will consider the following processes:

$$e + O_2(N_2) \rightarrow 2e + O_2^+(N_2^+),$$
 (1)

$$e + O_2 \rightarrow O^- + O, \tag{2}$$

$$O^- + O_2^+ (N_2^+) \to O + O_2 (N_2),$$
 (3)

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$$e + O_2^+(N_2^+) \rightarrow O(N) + O(N),$$
(4)

$$e + \mathcal{O}_2 \rightarrow 2\mathcal{O} + e, \tag{5}$$

$$O + O_2 + O_2(N_2) \rightarrow O_3 + O_2(N_2),$$
 (6)

$$O + O_3 \rightarrow 2O_2, \tag{7}$$

$$O^- + O_3 \to 2O_2 + e + 2.6 \text{ eV},$$
 (8)

$$O^- + O_2 \to O_3 + e - 0.42 \text{ eV}.$$
 (9)

With consideration of these processes the balance equation for the density of electrons N_e and neutral ions N has the following form:

$$\frac{dN_e}{dt} = (v_1 - v_2) N_e - \alpha_4 N_e N_+ + k_8 N_- [O_3] + k_9 N_- [O_2],$$
(10a)

$$\frac{dN_{-}}{dt} = v_2 N_e - \alpha_3 N_{-} N_{+} - k_8 N_{-} [O_3] - k_9 N_{-} [O_2].$$
(10b)

Here the subscripts denote the rate constants for frequencies of processes noted above, N_+ being the density of positive ions, while [X] is the density of the component X. Under the conditions to be considered the characteristic times for change in N_e , $N_$ significantly exceed the characteristic times for completion of the above elementary processes. This allows us to commence from quasisteady state solutions of system (10) in studying breakdown conditions, treating the slowly changing ozone concentration as a parameter. For the initial stage of the prebreakdown process ($N_e << N_-, N_- = N_+$) the quasisteady solution of Eq. (10) has the form

$$N_{-} = \frac{(\mathbf{v}_{8} - \mathbf{v}_{9})\mathbf{v}_{1}}{(\mathbf{v}_{2} - \mathbf{v}_{1})\mathbf{\alpha}_{3}}; \quad N_{e} = \frac{(\mathbf{v}_{8} - \mathbf{v}_{9})^{2}\mathbf{v}_{1}}{(\mathbf{v}_{2} - \mathbf{v}_{1})^{2}\mathbf{\alpha}_{3}}.$$
 (11)

As follows from Eq. (11), the equality of ionization and attachment frequencies ($v_1 = v_2$) implies increase without limit in the charged particle density, which can be considered as breakdown. However even at lower electric field intensities free electrons formed by the processes of Eqs. (8), (9) produce an accumulation of ozone which in turn stimulates further destruction of negative ions. This is the mechanism for reduction in the electrical strength of the air.

The equation for the ozone concentration has the following form:

$$\frac{d[O_3]}{dt} = M - [O_3]/\tau + k_5 N_e [O_2].$$
(12)

Here M is the volume ozone formation rate in the atmosphere in the absence of electric field; τ is the ozone molecule lifetime in the real atmosphere. The background ozone concentration in the real atmosphere $[O_3]_0$ is given by the expression

$$[\mathcal{O}_3]_0 = M\tau. \tag{13}$$

A typical value of this parameter is $[O_3]_0 \approx 10^{12} \text{ cm}^{-3}$. This allows us to neglect the process of Eq. (9) as compared to that of Eq. (8), with consideration of Eq. (11) writing Eq. (12) in the form

$$\frac{d[O_3]}{dt} = M + k_5 N_e [O_2] - \frac{(v_2 - v_1)}{k_8 \tau} (\alpha_3 N_e / v_1)^{1/2}.$$
(14)

It can easily be shown that a quasisteady solution of this equation exists for the condition

$$M\tau^{2} < (v_{2} - v_{1})^{2} \alpha_{3} / \{4k_{5} [O_{2}] v_{1} k_{8}^{2}\}.$$
⁽¹⁵⁾

TABLE 1. Proce	ss Rates in	n Atmosp	oheric Air
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<i>E N</i> , Td	20	40	60	80
v_1 , sec ⁻¹	$7,9.10^{-5} \\3,4.10^{3} \\1,6.10^{7} \\3,25$	2,8.10 ³	1,1.10 ⁶	$3, 5 \cdot 10^7$
v_2 , sec ⁻¹		2,3.10 ⁶	1,8.10 ⁷	$4, 2 \cdot 10^7$
v_5 , sec ⁻¹		6,5.10 ⁷	1,3.10 ⁸	$3, 0 \cdot 10^8$
v_9 , sec ⁻¹		175	6000	55000

TABLE 2. Atmospheric Parameters at Threshold of Instability

Parameter	<i>E/N</i> , Td				
	20	4ú	60	80	
N_, cm ⁻³	3,5	2,9.105	1,9.108	1,4.1011	
N_e , cm ⁻³	0,31	60	7,0.104	1,1.109	
τ , sec	4,2.104	160	12	0,026	

This condition, being a relationship between the critical electric field intensity and the background volume ozone formation rate M, defines the threshold for development of kinetic instability. This instability is accompanied by an increase in ozone and charged particle concentrations, and the condition under which it develops, Eq. (15), can be considered to be a breakdown criterion.

To obtain quantitative breakdown conditions we need data on the parameters of the elementary processes described in Eqs. (1)-(9). The limited reliability of available data will define the reliability of the present study's results. According to the measurements of [1], $k_8 = (3 \pm 1) \cdot 10^{-10}$ cm³/sec. It has been established reliably that $k_3 = 2 \cdot 10^{-6}$ cm³/sec. The value of k_9 , which depends strongly on electric field intensity, has been established by direct measurements of the process section [2]. To find the electric field intensity dependence of the rate constants k_1 and k_2 the numerical methods of [3] were used, with results corrected considering the equality of those constants at E = 25.5 kV/cm. The dependence of the O₂ molecule dissociation constant for electron collision upon electric field intensity [4] can be approximated as follows:

$$k_5 = 4 \cdot 10^{-8} \exp\left[-30/(E/N)^{1/2}\right],\tag{16}$$

where the parameter E/N is expressed in Townsends, and the rate constant in cm³/sec.

Table 1 presents the values of elementary rate constants as functions of electric field intensity used in the calculations. Table 2 gives critical values of ozone molecule loss times τ , corresponding to various field intensities and a background ozone concentration $[O_3]_0 = 10^{12} \text{ cm}^{-3}$. The basic ozone destruction mechanism in the real atmosphere is controlled by collisions with impurity molecules and microparticles. Convective removal of ozone from the electrified region may also play a certain role. Thus the electrical strength of the atmosphere is determined by its state, in particular, the degree to which it is polluted.

It should be noted that together with the kinetic instability considered above, thermal instability caused by the severe temperature dependence of the rate constant k_9 [5] may also lead to breakdown.

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