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Fast breakdown phenomena in liquid dielectrics under pulsed voltage conditions

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Abstract. In this work, the model of anode-initiated electrical breakdown in water under the condition of conservation of short-range order in crystalline lattice is proposed. The model is based on cascade Auger-transitions of electrons in valence band of the dielectric. Within this model, the process of generation of the free carriers and the dependence of breakdown channel propagation velocity on gap distance are considered. Also, the estimation of change in conductivity of the breakdown channel during the electrical breakdown in several liquid dielectrics is made using the data on velocity of breakdown channel propagation for time intervals from 1 to 8 ns. It has been shown that the increase in conductivity level in perfluoroeicosane, distilled water and transformer oil begins within 2–4 ns after the start of the breakdown processes. During the time of the channel propagation the conductivity rises by ~10–30 % of the initial value. Probably, this rise in conductivity is associated with the impact ionization of the dielectric material at the boundaries of the breakdown channel by the conduction electrons.
Keywords: electrical breakdown, breakdown mechanism, breakdown in liquids, pulsed breakdown

1. Introduction

To increase the reliability of high-voltage devices such as pulse transformers, pulse forming lines of accelerators, liquid-based dischargers etc., it is very important to understand the fundamentals of insulator's electrical strength failure. Pulsed electrical breakdown is one of the ways by which the dielectric might come to such a failure and is one of the most interesting phenomena from the point of view of the pulsed power [1].

The works devoted to ionic composition of the breakdown products of solids may give some important information for understanding of nature of the same process in dielectric liquids. Particularly, the works on mass spectrometry of plasma bunches ejected in vacuum from the region of surface flashover and from the breakdown channels in solid dielectrics shows that the main part of the ionic charge is carried by the singly ionized molecules. In the case of KCl there are ions KCl^+, K_2Cl^+, $(K_2Cl_2)^+$ [2] and in the case of polyethylene [2, 3] there are singly ionized clusters $C_2H_5^+$. $C_4H_9^+$, $C_8H_{17}^+$ comprising of several monomers. The ionization degree in the breakdown channel in such cases doesn't exceed 10 %. It may be assumed, that the main fraction of neutral particles has the same composition. The presence of the gas phase is confirmed by the ejection of a bunch of ionized particles at the moment, when the breakdown channel tip reaches the free surface of the dielectric and the matter from the channel is released in vacuum [4].

By contrast to the solids, there are some differences in structural failure of dielectric liquids. Theoretical works show [5], that in strong electrical field the molecular chains of transformer oil are being broken by water ions and form carbon-containing components CH, CH_3, C_3H_5 and C_7H_6. The products of decomposition form the gap of electron conductivity in the forbidden gap of oil molecules, which might be considered as formation of the conducting layer. The understanding of how liquid dielectric start to degrade under strong electric field and the degree of this degradation in real time, as well as the possible ionization mechanism are of practical interest.

According to works [6–8], the anode-initiated electrical breakdown in liquid dielectrics might be described by the mechanism of cascade Auger transitions of electrons in valence band of dielectric. Initiation, growth and orientation of the breakdown channels are explained taking into the account the data on chemical and energetic structure of the considered materials. Particularly, this theoretical model works out well for liquid perfluoroeicosane [6]. One of the key features of this model is the explaining of the high velocities of breakdown channels propagation in crystals. In fact, some liquids

also demonstrate high breakdown velocities [6], which provide the reason to consider them within the crystal-oriented model. It was interesting to apply this model to structurally simple and widely used liquid as water, which among other applications may be used as insulator in high-voltage pulse forming lines [1].

In the present work, the process of anode-initiated electrical breakdown in liquid water is considered within the model which implies generation of free carriers by interatomic Auger transitions of electrons in valence band of dielectric. The accompanying phenomena are considered, e.g., the increase in conductivity level of the breakdown channel during the electrical breakdown. The estimation of such increase in several liquid dielectrics is made using the data on velocity of breakdown channel propagation for time intervals from 1 to 8 ns at voltages of up to 140 kV.

2. Model of breakdown channel formation in water

Computational models of hexagonal ice I_h are based on Bernal–Fowler rules: each oxygen atom is in the center of tetrahedron having four other oxygen atoms at its apices spaced apart at 2.76 Å. Along all the four nearest directions O–O, there are hydrogen atoms two of which form covalent bond O–H, the other two form hydrogen bond O···H with oxygen atom in the center. Each water molecule is connected by hydrogen bonds with 4 nearest neighbors. Such a molecule arrangement provides relatively large intermolecular attraction within the lattice. The lattice itself consists of layers which are perpendicular to the c-axis of the crystal and contain hexagonal rings formed by water molecules. There are also the hexagonal rings formed by three molecules from one layer and three molecules from the next layer (see Fig. 1) [9].

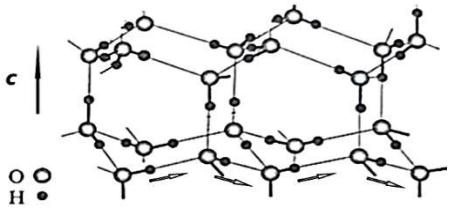

Fig. 1. Intermolecular bonds in hexagonal ice [9]. Direction of the breakdown channel propagation is shown by arrows.

Water molecules in ice I_h are slightly topologically different from the isolated ones. So, for ice the distance O–H is 1.01 Å (0.96 Å for isolated molecule), and the angle H–O–H, probably, a little bit bigger than the bond angle in the isolated molecule, which is 104.5° [10, 11]. Obviously, there is no long-range order in water. However, at short range the structure of water is topologically the same as that of I_h ice, but has flexible bonds and flexible angles within H_2O tetrahedrons [11].

The diagrams of energy bands in the strong electric field in the region of contact of metal electrode and water are presented at Fig. 2.

Fig. 2. Resonance (a) and Auger (b) transitions in water in strong electric field. W_F – Fermi level of metal, W_c – conduction band bottom energy level, W_v – valence band top energy level.

Energy states of hydrogen and oxygen atoms are given according to data from [10, 11]. According to the assumed designations [10, 11], the energy states of liquid water $1b_1$ form the top of the valence band. These states are of 2p-type. The electron density, corresponding to this band, is localized in oxygen atoms. H-bond includes hybridization of $3a_1$-orbitals (where $3a_1$ – eigenstates of water O 2p hybridized with H 1s) with $1b_1$-orbitals, that results in formation of a new hybrid state $3a_1/1b_1$. Density of states in the bottom of the conduction band is formed by 2p/3s-states of oxygen and a share of 1s-states of hydrogen.

According to [6–8], beginning of the formation of the breakdown channel is conditioned by the processes at the metal – dielectric boundary. According to rough estimation, the average strength of electric field in the dielectric placed between the electrodes is $\sim 10^6$ V/cm. However, the maximum value of the local electric field may be much bigger because of the presence of microprotrusions on the electrodes, which enhance the field up to $\sim 10^8$ V/cm and higher.

In the near-electrode region the electrons tunnel into metal, which results in formation of two ions O^{2+} with two holes at $1b_1$-orbitals. To make this tunneling possible, it is necessary to provide voltage U, that is sufficient to rise the top edge of valence band up to Fermi level of the metal (Fig. 2a). Probably, the recombination of the hole occurs due to resonance transition of the electron from $3a_1/1b_1$- band of hydrogen to $1b_1$- band of oxygen. This process is possible if the energy of the $3a_1/1b_1$-band is raised by ~3 eV (Fig. 2a). Decay of the hole in H^+ occurs due to interatomic transition of Auger electron from $1b_1$-orbital of neighboring oxygen atom and consequent injection of Auger electron into the conduction band (Fig. 2b).

The transition of Auger electron in the conduction band occurs provided that minimal energy gap between $3a_1/1b_1$-orbitals of H^+ and $1b_1$-orbitals of O in electric field not less than the band gap of the dielectric. The questions concerning the estimation of electric field strength at the breakdown channel front are considered in work [8]. The necessary band bending is provided by the electric field of the volume charge.

A single cycle of the volume charge propagation may be decomposed into three consecutive stages. At the first stage, when the critical strength of electric field ($\sim 10^8$ V/cm) is reached, the electron transfers from hydrogen atom to O^{2+} by the resonance transition. Thus, the breakdown channel propagates at the distance Δx_1. The duration of this stage is $\tau_r \sim 10^{-16}$ s.

At the second stage, the electron from oxygen atom transfers to H^+ due to interatomic Auger transition in $\tau_A \sim 10^{-16}$ s. As a result, the hole decays and an Auger electron is injected into the conduction band of the dielectric. In this way, the breakdown channel propagates at one more interatomic distance Δx_2 (Fig. 2b). Negative charge of electrons weakens the field of holes and hence weakens the band bending near the volume charge boundary.

At the third stage, the electrons are pulled out from the region of the volume charge by the external electric field. Thereby, the critical electric field strength is reached for the next cycle to be executed. The front of the breakdown channel coincides with the volume charge boundary. Time of the cycle Δt can be represented as

$$\Delta t = \tau_r + \tau_A + \tau_1, \tag{1}$$

where τ_1 is time to reach the critical electric field strength.

The breakdown channel is formed by the bunch of single channels of Auger transitions (Fig. 1) which go in one direction. Structurally, the breakdown channel consists of a region of volume charge and a conducting region which contains the electron-hole plasma. The time τ_1 can be calculated as

$$\tau_1 = Q_A / i_1, \tag{2}$$

where Q_A is the charge density of Auger electrons, i_1 – the conduction current density.

Formula (1) allows us to estimate the dependence of velocity of the breakdown channel propagation in the interelectrode gap as $v \approx (\Delta x_1+\Delta x_2)/\Delta t$.

3. Experimental results and discussion

The measurements of time to breakdown t_b in perfluoroeicosane, distilled water and transformer oil in the range of interelectrode gaps of $d \sim 0.1$–1 mm are performed in point-to-plane electrode configuration [6, 12].

According to the calculations in [8], the velocity of the breakdown channel propagation $v = d/t_b$ is proportional to $E_{mid}\cdot\sigma^2$, where $E_{mid} = U/d$ is the average strength of external electric field in the sample and σ is the conductivity in the breakdown channel. Experimental values of velocity v in different dielectrics are presented at Fig. 3. The obtained velocities decrease as the field strength decreases in wider gaps. The character of this dependence is close to hyperbolic. Theoretical curves of change in velocity of breakdown channel propagation $v_T \sim 1/d$ (at fixed value of the initial conductivity σ_0) are shown by dashed lines. In works [8, 12], the connection between velocity of breakdown channel propagation and conductivity is defined by the equation:

$$\frac{v}{v_T} \approx \left(\frac{\sigma}{\sigma_0}\right)^2. \tag{3}$$

Values of relative conductivity σ/ σ_0 of the breakdown channel for the tested materials are presented at Fig. 3.

Increase in conductivity level occurs in ~2–4 ns after the beginning of channel growth. The conductivity increases by ~10% of the initial value during the time of channel propagation. Heating of the material in the channel and increase in pressure are connected with the process of recombination of electrons and holes.

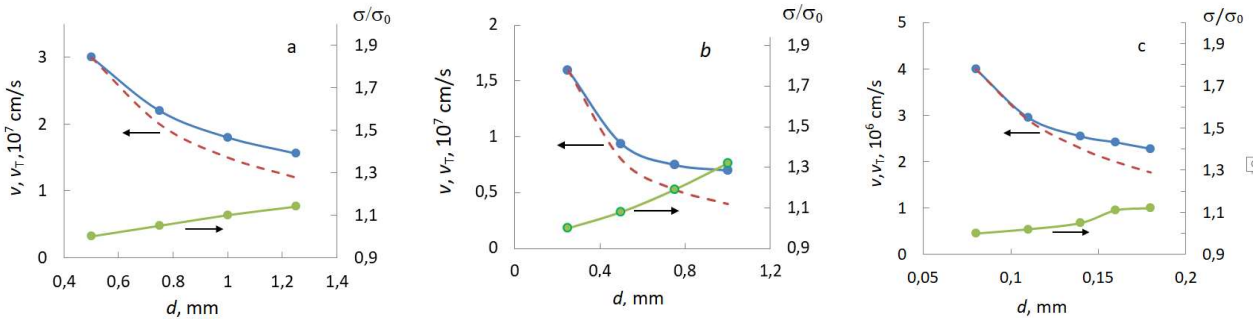

Fig. 3. The dependence of velocity of breakdown channel propagation v and relative conductivity σ/σ_0 on the electrode gap in liquids: *a* – perfluoroeicosane, *b* – distilled water, *c* – transformer oil.

The concentration of electrons, probably, decrease from $n_1 \sim 10^{21}$ down to $n_2 \sim 10^{16}$ cm^{-3} [8]. Thermal energy is equal to $W_T = (n_1–n_2)W_g$, where $W_g = 9$ eV is the band gap for water. Value of W_T for water is 1.44 kJ·cm^{-3} and value of thermal capacity c_p is 4.18 J/g·K, so the temperature of heating may be calculated as $\Delta T = W_T/c_p \approx 340$ K. Thus, we can distinguish two different regions within the breakdown channel, which are vaporized region (likely, with molecules H_2O, ions H_2O^+ and electrons) and the heated liquid region at the channel front where the recombination of volume charge with charge carriers takes place. It is interesting to reveal the basics of the consequent conductivity rise. Although the electric field strength drops as d increases, it remains ~10^6 V·cm in the area of interest. High values of the additional conductivity might indicate the impact character of charge carriers generation.

Ionization of isolated H_2O molecule by the electron impact demands energy W_i of ~13 eV. At the strength of electrical field $E_{mid} \sim 10^6$ V/cm, the electron gains energy W_i (without losses) at a distance $l = W_i/eE_{mid} \approx 1.3$ μm. The component of electric field which is normal to the breakdown channel surface and the presence of positively charged water molecules on the inner surface of the channel reduce the work function.

Probably, the increase in mobility of electrons in vaporized environment allows them to achieve the energy of ionization of the material at the channel boundaries. An evidence for this hypothesis is the fact, that in alkali-halide crystals the breakdown channel diameter near the positive electrode increases as the channel grows [13].

The results on pre-discharge current in water [14] speak in favor of the impact ionization mechanism. The time dependence of the current during the anode-initiated in a 1-cm layer of water is presented at Fig. 4.

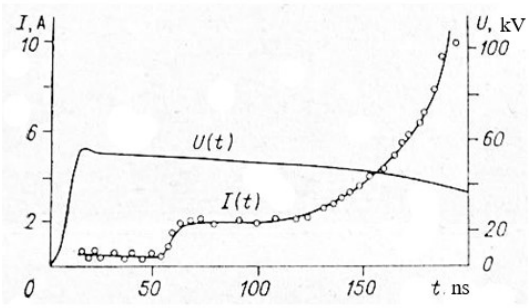

Fig. 4. Pre-discharge current in water and voltage across the measuring capacitor [14].

The rise of current at ~50 ns corresponds to the beginning of the channel growth from the needle. At the initial stages of the expanding of the conduction area in the bulk of the sample, the current remains stable (~2 A) until the area expands to ~5 mm, i. e. approximately half of a distance from the tip of the needle electrode to the plane electrode. After that, the current increases by several times before the conducting channel spans the interelectrode gap. At the average electrical field strength of $\sim 5 \cdot 10^4$ V/cm, velocity of breakdown channel front propagation is $\sim 6.7 \cdot 10^6$ cm/s.

As it is shown in work [8], the amplitude of pre-discharge current may rise not more than few percent as the channel grows though the sample. Superlinear increase in concentration of nonequilibrium carriers (Fig. 4) is associated with the secondary processes in the breakdown channel. A specific feature of this current curve is the periodical doubling of current every $\tau_i \approx 16.6$ ns (within the interval of ~1.1 mm), which corresponds to the impact ionization of material by the fast electrons. If we assume the time of the breakdown channel front as t which meets the requirement $0 < t < t_b$, then the increase in current (Fig. 4) may be represented as an exponent $I_i(t) \sim 2^n$, where $n = t/\tau_i$ is the whole number.

4. Conclusion

It has been shown that the electrical breakdown in distilled water may be described by the model of cascade Auger transitions in the valence band of the dielectric under the condition of conservation of short-range crystalline order.

In perfluoroeicosane, distilled water and transformer oil the increase in conductivity level by ~10–30 % of the initial value during the propagation of the breakdown channel is observed. Probably, such an increase has a relation with the destruction of the dielectric material at the breakdown channel boundaries. The relation between the change in conductivity of the breakdown channel and velocity of the channel propagation is considered for the nanosecond range of high-voltage pulses.

The analysis of the structure of the pre-discharge current during the sub-microsecond breakdown in water shows that superlinear periodical doubling of nonequillibrium carriers during this process corresponds to avalanche generation of electrons. Predominant factor of the conductivity increase is the breakdown time duration t_b.

5. References

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