

## PECULIARITIES OF SPACE ELECTRIC CHARGE FORMATION IN A LIQUID

E.I. SKIBENKO, V.B. YUFEROV, I.V. BURAVILOV, YU.V. KOVTUN,  
A.N. PONOMARYOV

UDC 537.528  
©2008

National Scientific Center "Kharkiv Institute of Physics and Technology"  
(1, Akademichna Str., Kharkiv 61108, Ukraine; e-mail: Ykovtun@kipt.kharkov.ua)

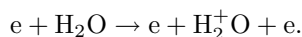
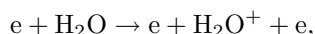
The paper presents experimental and theoretical data on the water medium ionization by a high-voltage high-current pulsed discharge. Physical mechanisms of ionization at the linear and exponential stages of the discharge are discussed.

According to the literature data [1], there are two basically different approaches to the phenomenon of electric discharge in a liquid. By the first variant, an electric discharge in a liquid is considered as a certain analog of the gas cavity discharge. By the second variant, a discharge in a liquid is considered as a consequence of the avalanche-like multiplication of free charge carriers in the liquid itself. By this model, the electrons can be accelerated in intense fields in a liquid and ionize molecules and atoms. There is also a hypothesis that, by the ionization mechanism of breakdown in liquids, the initial electrons provoking the impact ionization are generated in a liquid due to the cathode field electron emission. At the same time, the impact ionization has not been convincingly confirmed experimentally as a base of the electric mechanism of the breakdown in liquids.

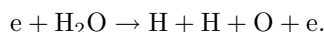
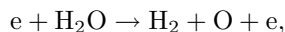
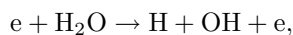
In the present work, the authors made an attempt to determine experimentally the electron density of a high-current high-voltage pulsed discharge plasma being formed in the water medium. The data obtained are compared with the data calculated in terms of the effective cross-sections of the ionization by electron impact.

The nomenclature of participating particles formed as a result of elementary processes at the initial discharge stage includes the following molecules, atoms, and ions:  $\text{H}_2\text{O}$ ,  $\text{H}_2$ ,  $\text{O}_2$ ,  $\text{OH}$ ,  $\text{H}$ ,  $\text{O}$ ,  $\text{O}^+$ ,  $\text{H}^+$ ,  $\text{H}_2^+$ ,  $\text{O}_2^+$ ,  $\text{OH}^-$  *etc.* Among diverse probable types of collisions, the following basic elementary processes of atomic and electron collisions should be mentioned [2]: ionization and excitation by electron impact, dissociation and dissociative ionization by electron impact, and ion-atom exchange. These collisional processes can be written in the following form:

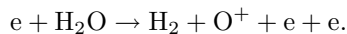
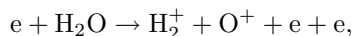
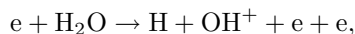
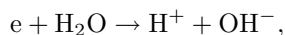
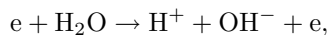
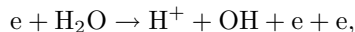
### 1. Ionization by electron impact



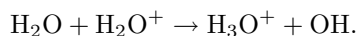
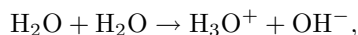
### 2. Dissociation by electron impact



### 3. Dissociative ionization by electron impact



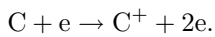
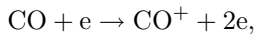
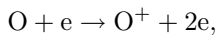
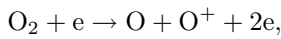
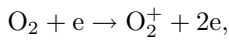
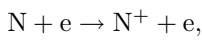
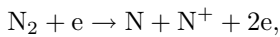
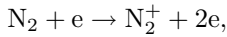
### 4. Ion-atom exchange



It should be noted that, at electron energies of 50 eV or more, the effective ionization cross-section and dissociative ionization cross-section of a given polyatomic molecule differ slightly from each other. Therefore, in real calculations and estimations, one can

use one of these values taking into account both of the ionization channels.

To complete the picture of the ionization process, one should consider also the gases dissolved in a liquid. Among these are  $N_2$ ,  $O_2$ ,  $CO_2$ . The processes of collisions with participation of  $N_2$ ,  $N$ ,  $O_2$ ,  $O$ ,  $CO_2$ , and  $CO$  particles can be written as follows:



The vapor-gas fraction of the water medium can be ionized by two groups of electrons: a low-energy one with the energy of an order of several tens of eV and a high-energy one with the energy in the keV range. The electron energy range in the first case (low energies) was given with taking into account the fact that, by assumption, the main source of initial electrons in the discharge is the field emission of electrons from the cathode.

In the first case (low energies), the value of the effective ionization cross-section is in the range  $10^{-18}$  –  $10^{-16} \text{cm}^{-2}$ , and, in the second case (high energies), it is of the order of  $10^{-18} \text{cm}^{-2}$ . The probable presence of two groups of electrons – thermal and accelerated ones, is related, on the one hand, with the field emission of electrons from the cathode surface and, on the other hand, with the existence of electric field strengths of the order of  $10^5 \text{V/cm}$  in the discharge [3].

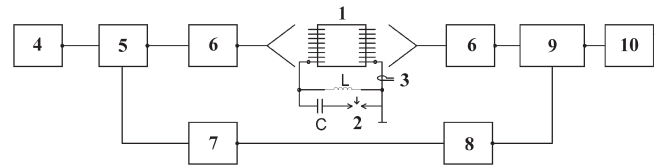


Fig. 1. Block diagram of the experimental device for plasma density measurements. Symbols: 1 – discharge gap, 2 – discharger, 3 – Rogowski loop, 4 – microwave oscillator, 5 – coupler, 6 – antenna, 7 – attenuator, 8 – phase shifter, 9 – measuring microwave diode, 10 – recording unit, C – single capacity storage, L – inductance

Experiments were carried out at the installation (Fig. 1) comprising a discharge cell of  $590 \text{cm}^3$  in volume, a pulsed multichannel power-supply system, and diagnostic facilities [4]. The eight-electrode discharge gap was 2.3 cm in width and 16 cm in length. For the diagnostics of a plasma being formed, the method of microwave sounding was selected, which allowed us to measure directly the electron component density, and a microwave interferometer with the operating wavelength  $\lambda = 0.8 \text{cm}$  was used as a measuring device. The discharge current value was measured by means of the Rogowski loop.

In experiments, the interferogram was measured, i.e. the phase shift value (phase incursion)  $\Phi/2\pi$  of the microwave signal changing in time from the initiation of a discharge to its end. Then, using the equation

$$\frac{\Phi}{2\pi} = \frac{L}{\lambda} \left( 1 - \sqrt{1 - \frac{n}{n_{cr}}} \right) = \frac{nL}{2\lambda n_{cr}}, \quad (1)$$

it is possible to determine the plasma density  $n$  at a given time instant. Here,  $\lambda$  – operating wave length;  $n_{cr}$  – critical plasma density for the given wave length;  $L$  – transverse size of the plasma formation, and  $nL$  – its thickness in  $\text{cm}^{-2}$ . At the time instant of the gas-vapor phase formation and during its existence, the attenuation coefficient of a microwave signal was from 0.3 to 3 dB, which was in good agreement with the literature data [5–6].

Figure 2, *a* presents the density arising in a plasma discharge as a function of the time. The present dependence can be conditionally divided into three parts.

The first is a part of the plasma density increasing during  $12.5 \mu\text{s}$ . The next is a part, where the maximum values of  $nL$  are reached and some stationary conditions can exist during  $18 \mu\text{s}$ . The third part shows the plasma formation thickness decrease and the plasma formation decay during  $20 \mu\text{s}$ . The measured time balance allows us to affirm that the duration of the preionization and ionization stages is near  $50 \mu\text{s}$ .

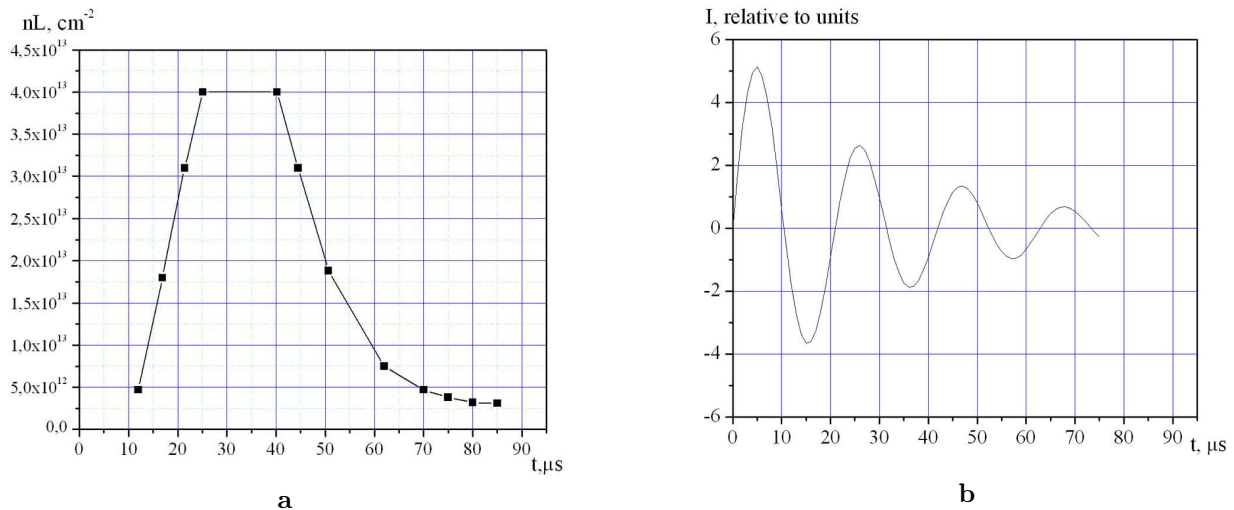


Fig. 2. Plasma density (a) and the discharge current (b) as functions of time

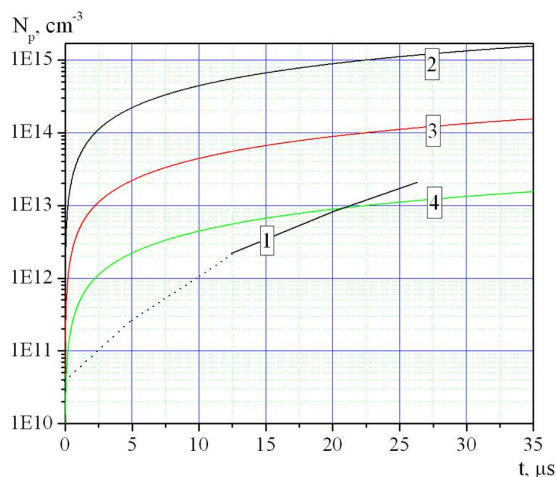


Fig. 3. Comparison of experimental data with the calculated results. 1 – experiment; 2,3,4 – results of calculations at electron energies of 80 eV, 5 keV, and 30 keV

Figure 2,b presents the discharge current as a function of the time. It is seen that the plasma density decrease begins, as the density decreases approximately by 50% of the nominal and more.

When the electron flow of a density  $n_e$  and a rate  $v_e$  passes through the neutral medium (for example, a water vapor and gases dissolved in it) with a density  $n_0$ , then the plasma is formed having a density  $N_p$  that is determined by the expression

$$N_p = n_e v_e \sigma_e n_0 \tau, \tag{2}$$

where  $\sigma_e$  is the cross-section of the neutral gas ionization by electrons [7];  $\tau$  is the plasma lifetime.

If we take the value of  $\tau$  equal to the experimentally determined plasma lifetime (flight time), we obtain the value of the equilibrium density of the plasma formed by the electron flow. In Fig. 3, curves 2–4 present the time dependences of the plasma density formed due to the collisional (impact) ionization calculated by formula (2) for different electron flow energies.

It is seen that the density of the plasma formed exceeds the value of the primary electron beam density by an order of magnitude.

Thus, in future, the ionization of a neutral gas-vapor medium can be carried out, in main, not by the primary beam electrons, but by the plasma electrons accelerated due to collective interactions [8–9]. In this case, the density change is determined by the equation

$$dN = \langle \sigma_e v_e \rangle n_0 N_p dt, \tag{3}$$

where  $N_p$  is the current plasma density.

Extrapolation of the experimental curve  $N_p = f(t)$  into the region of low plasma density values up to the intersection with the similar calculated curve gives the value of the starting plasma density, after reaching which the exponential increase in the plasma density to about  $3 \times 10^{10} \text{ cm}^{-3}$  begins.

The use of the relation  $\langle \sigma_e v_e \rangle = f(E_e)$  and the comparison of literature data with experimental one (see Fig. 4) allow us to evaluate the velocity (energy) of plasma electrons ionizing the medium at an initial stage of the discharge.

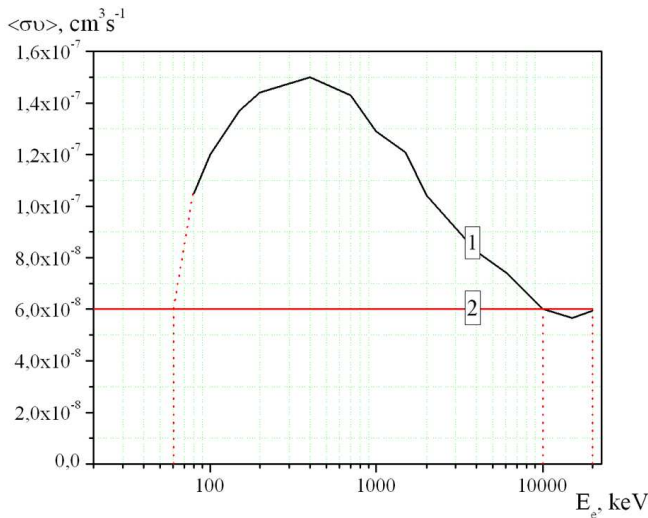


Fig. 4. Average product  $\langle \sigma v \rangle$  as a function of the electron energy: 1 – literature data, 2 – experiment

This estimate gives the following values of  $E_e$ : near 60 eV in the low-energy part of the spectrum and 10–20 keV in the high-energy part.

The current density of the electron beam (flow) ionizing the water medium during a high-voltage electric discharge was calculated, at the stage of the exponential increase of the density of the plasma formed, by the equation [8]

$$L \approx 10^{-8} \frac{E_e}{j} \sqrt{n_p}, \quad (4)$$

where  $L$  is the beam-plasma interaction length, cm;  $E_e$  is the beam electron energy, eV;  $j$  is the beam current density, A/cm<sup>2</sup>; and  $n_p$  is the plasma density, cm<sup>-3</sup>.

As follows from Fig. 4, the energy of electrons ionizing the water medium can take the following values: it is 60–100 eV in the low-energy part of the spectrum and 10–20 keV in the high-energy part. In Fig. 5, we give the values of a minimum electron beam (flow) current density, at which the beam-plasma interaction (BPI) is realized in the medium under study with the geometry as mentioned above including the interaction length  $L = 2.3$  cm and the plasma density value  $n_p = 3 \times 10^{10}$  cm<sup>-3</sup> obtained in the experiment, at which its exponential increase begins. A maximum summary electron current in a discharge can reach the value  $I_\Sigma = j = 550$  A, and its minimum value is 1.6 A. The value of  $\gamma$  of the amplitude rise of longitudinal high-frequency oscillations with frequencies close to the Langmuir frequency [8] is in the range from  $1.5 \times 10^8$  to  $2.5 \times 10^9$  s<sup>-1</sup>.

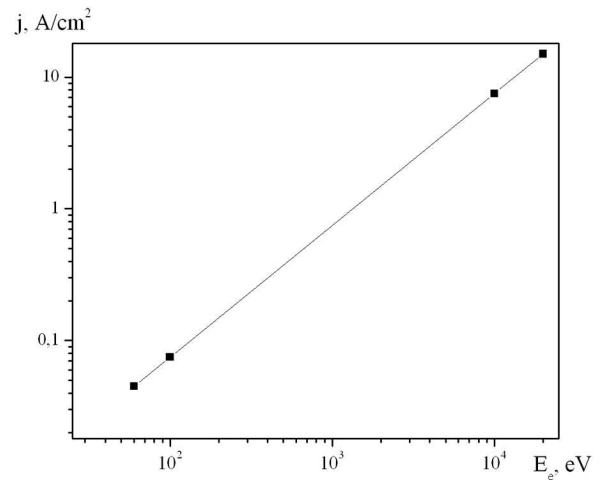


Fig. 5. Calculated dependence of the minimum value of the electron beam (flow) current density, at which BPI is realized, on the beam energy, the range of its change being determined experimentally (see Fig. 3)

The estimation, similarly to [10], gives the value of the specific energy expenditure for the plasma formation of  $(2 \div 4) \times 10^{-2}$  J/cm<sup>3</sup>. The total energy expenditure throughout the volume of 600 cm<sup>3</sup> can reach 25 J.

## 1. Conclusions

For the first time in practice of investigations on electrical discharges in a liquid media, the measurement of discharge microparameters has been carried out using the microwave methods of plasma diagnostics, in particular, the plasma density and its dynamics in time.

The experimentally measured rate of increase in the plasma density at the initial stage of a discharge is comparable with the density increase in the case of the ionization by the electron impact and thus can evidence the ionization mechanism of breakdown in the liquid medium, when the pulsed voltage is applied.

It is shown that a high-voltage mean-intensity breakdown in a liquid medium under a spatially distributed discharge can be realized at a current pulse duration up to 100  $\mu$ s.

The result obtained differs from the literature data [11], where strongly compressed discharges with transverse dimensions of 10–30  $\mu$ m were studied. In our case, we consider a spatially distributed discharge having an actively ionized volume of 100–300 cm<sup>3</sup> with an effective volume of the discharge cell of  $\sim 600$  cm<sup>3</sup>.

The discharge development in a liquid can be conditionally divided into several stages: the thermal

stage, when the energy contribution to the discharge is spent in the process of local vapor formation; the ionization stage, when a part of the energy is spent for the formation of free electrons and, respectively, negatively charged clusters; and, at last, the stage of plasma balance maintaining during the discharge current flowing.

The composition of particles participating and generated as a result of the elementary processes in the initial – ionization – stage of a discharge is determined. The main elementary processes taking place in the electric charge in a liquid medium are: ionization and excitation by electron impact, dissociation and dissociative ionization by electron impact, and ion-atom exchange.

The plasma density was determined using a microwave interferometer on the wave length  $\lambda = 8$  mm. The measured plasma density value is  $1.8 \times 10^{13} \text{ cm}^{-3}$  and more. By the same technique, the dynamics of an increase in the plasma density and a decrease in the discharge was determined. For the total discharge duration of  $100 \mu\text{s}$ , the increase in the plasma density by the order of magnitude from  $2 \times 10^{12}$  to  $2 \times 10^{13} \text{ cm}^{-3}$  is observed during  $12.5 \mu\text{s}$ . Further, the maximum density values are reached and some stationary conditions can exist during  $18 \mu\text{s}$ . The decrease in the plasma density and the plasma formation decay have the total duration of  $20 \mu\text{s}$ . The measured time balance allows us to affirm that the duration of the preionization and ionization stages is near  $50 \mu\text{s}$ .

The presence of the stage of a linear increase in the plasma density due to the impact ionization by primary electrons and the stage of the exponential increase in the plasma density due to the accelerated plasma electrons as a result of collective interactions was observed. The starting plasma density necessary for its exponential increase is of about  $3 \times 10^{10} \text{ cm}^{-3}$ .

Ionization of the gas-vapor fraction of the water medium is caused by two groups of electrons: low-energy and high-energy ones. The electron energy of the low-energy part of the spectrum is of 60 eV and that of the high-energy one is 10–20 keV. The estimation of the energy expenditure for the water medium ionization gives the specific energy expenditure for the plasma formation of  $(2 \div 4) \times 10^{-2} \text{ J/cm}^3$ . The total energy

expenditure throughout the volume of  $600 \text{ cm}^3$  can reach 25 J at the total energy contribution of 100 J. The ionization expenditure in a liquid is several times higher than that in a discharged gas. The stages of electric discharge development in a liquid can be represented in the following way: gas-vapor phase formation – breakdown – gas-vapor phase dissolution in the liquid.

1. V.Ya. Ushakov, *Pulse Electrical Breakdown in Liquids* (TSU, Tomsk, 1975) (in Russian).
2. J.B. Hasted, *Physics of Atomic Collisions* (Elsevier, New York, 1972).
3. *Encyclopedia of Low-Temperature Plasma*, edited by V.E. Fortov (Nauka, Moscow, 2000) (in Russian).
4. E.I. Skibenko, V.B. Yuferov, I.V. Buravilov, and A.N. Ponomarev, *Zh. Tekhn. Fiz.* **76**, 133 (2006).
5. V.Yu. Katkov, V.Ya. Ryadov, N.I. Furashov, in *Abstracts of reports at the III All-Union Symposium on Millimeter and Submillimeter Waves, 1980*, p. 287 (in Russian).
6. Yu.S. Bashkin, E.V. Sukhonin, and V.I. Chernyshov, in *Abstracts of reports at the III All-Union Symposium on Millimeter and Submillimeter Waves, 1980*, p. 271 (in Russian).
7. C.F. Barnett, J.A. Ray, E. Ricci, M.I. Wilker, McDaniel, E.W. Thomas, and H.B. Gilbody, *Atomic Data For Controlled Fusion Research* (Oak Ridge National Laboratory, USA, 1977).
8. Ya.B. Fainberg, *Atom. Energ.* **11**, 313 (1961).
9. I.F. Kharchenko, Ya.B. Fainberg, R.M. Nikolaev, E.A. Kornilov, E.I. Lutsenko, and N.S. Pedenko, *Yad. Sintez, Appl.*, **3**, 1101 (1962).
10. E.A. Lysenko, E.I. Skibenko, and V.B. Yuferov, *Influence of Vacuum Conditions on the Supply of Light Admixtures to Plasma on the Initial Stage of a Discharge: Survey* (TsNII Atominform, Moscow, 1987) (in Russian).
11. L.A. Yutkin, *Electrohydraulic Effect and Its Applications in Industry* (Mashinostroenie, Leningrad, 1986) (in Russian).

#### ОСОБЛИВОСТІ УТВОРЕННЯ ОБ'ЄМНОГО ЕЛЕКТРИЧНОГО РОЗРЯДУ В РІДИНІ

Є.І. Скібенко, В.В. Юферов, І.В. Буравілов, Ю.В. Ковтун, О.М. Пономарьов

#### Резюме

Наведено експериментальні і розрахункові дані з іонізації водного середовища висковольтним потужноструммовим імпульсним розрядом. Обговорюються фізичні механізми іонізаційного процесу на лінійній і експонентній стадіях розряду.