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Self-running low-frequency pulsed regime of DC electric discharge in gas bubble immersed in a liquid

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The paper presents the set of experimental data on periodical electric breakdown inside a single gas bubble inserted in narrow dielectric tube filled with a liquid. Each electric breakdown is accompanied with slow extension of the bubble and decreasing the current. Once a length of the bubble reaches the critical magnitude, the current falls down and the bubble shrinks quickly to its initial size. Thereafter new breakdown appears again. A simplified model of the phenomenon observed is described as well.

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1. Introduction

Effective generation of bio-chemically active but environmentally friendly species like O, OH, etc inside a liquid is of great interest for many science and practical applications (water purification, biomedical applications, etc). In many cases, short-pulsed discharges forming the streamers in bulk of a liquid or in gas above liquid surface are used to do that [1-5].

Another promising approach allowing us to generate the radicals in abundantly within the liquid is an electric discharge in the water filled with chaotically moving gas bubbles [6-9]. Under some conditions, gas discharge occurs in the bubbles. In such a case, active species have been produced by cold plasma inside small bubbles (but not in a liquid itself), and after that they are transported due to diffusion from the bubbles into the liquid.

Taking this in mind, an elaboration of deep insight into the mechanism of the gas discharge formation inside the bubbles is one of the important tasks. Comprehensive knowledge on this topic will help to find out the optimal regimes for plasma activation of a liquid.

For normal situation, gas bubbles are not located at fixed points in a liquid but they always travel chaotically and rise to the top due to Archimedes' force. This circumstance (i.e. continuous changing the bubble position) hampers seriously the experimental investigation on dynamics of an electric breakdown development in a single gas bubble.

Besides, breakdown voltage $U_b=E_b l$ across bubble surrounded by conductive liquid can be reached only under pulsed superposition of high voltage U_0 on the liquid. Here E_b is the gas breakdown electric field strength, l is the length of a bubble. A proportion $U_0>U_b$ is the needed condition providing fast breakdown development [10].

To provide the term mentioned above, a typical rise-time τ of the pulse has to meet the second condition: $\tau<\varepsilon\varepsilon_0/\sigma$ (in other words, displacement current in a liquid has to exceed conduction current during pulse rise time). Here ε and ε_0 are dielectric permittivities of the liquid and vacuum.

So, for the case of high magnitudes in both a conductivity σ of the liquid and gas breakdown electric field strength U_b, the sophisticated and expensive pulsed generators forming very short (in nanosecond range) and very high (many kilovolts in amplitude) pulses are required.

2. The experimental set-up

To avoid two serious problems mentioned above (bubble chaotic emersion and using of nanosecond region high-voltage pulses), we have done the experiments under conditions allowing permanent localization the bubble(s) at fixed points. To do it, narrow (inner diameter is 2.5 mm) and long (the length is 100 mm) quartz tube oriented horizontally was used (see Fig.1). Our set up allows us to make experiments with several bubbles simultaneously but the experiments presented here were performed with a single bubble.

Quartz tube was filled with water solution having different electric conductivity (20 μS/cm for distilled water and 12 mS/cm for physiological solution). Initial bubble of 2.5 mm in a diameter and 4-5mm in the length was filled with ambient air at atmospheric pressure. It is a crucial point that bubble length exceeds the tube diameter. Namely this circumstance allows us to avoid strong requirements on the pulse rise-time imposed by the second condition: $\tau<\varepsilon\varepsilon_0/\sigma$.

Initial state of the wall in quartz tube is hydrophobic, and there is no the wet inner wall of the capillary occupied by bubble. In other words, there is no thin layer of a liquid on the wall shunting the poles of a bubble, and a conduction current passes therefore only through plasma channel produced by gas discharge within a bubble. However after discharge activity inside the tube over period of tens seconds (or longer –it depends on type of liquid), inner wall can be hydrophilic. It results in drastic change in behavior of the breakdown. We took this circumstance into account, and all our experiments were done therefore with discharge pulses which have duration only 3 s.

High-voltage pulses up to 20 kV in the amplitude were applied to the HV terminal. Typical rise-time and duration of the pulses are 0.1μs and 3s respectively. The recording of the current and voltage waveforms was done by a TDS520D oscilloscope. The pictures of bubbles with a gas discharge were taken by a Panasonic NV-GS500 video camera with exposure time of 20 ms.

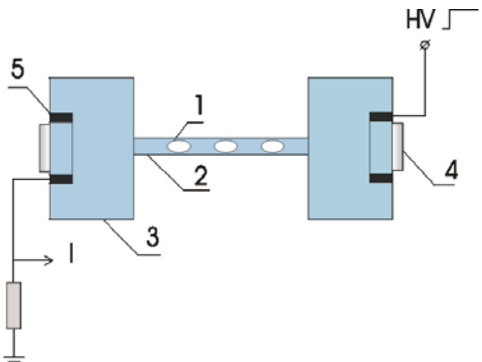

Fig.1. Sketch of the experimental set up. 1 is the gas bubble; 2 is the quartz tube; 3 is the water reservoir containing metallic electrode 5; the water in reservoirs was kept always at atmospheric pressure; 4 is the quartz window; HV is high-voltage terminal connected with power supply through resistor of 158 kOhm.

3. Results of the basic experiments

We have revealed that electric breakdown in a single gas bubble inserted in the narrow dielectric tube filled with a conductive liquid exhibits itself in a filamentary mode (see Fig.2). An individual thickness of these numerous filaments is smaller compared with an inner diameter of the tube. The filaments are non-stationary in the space and time, twist like snakes and therefore non-uniformly occupy the bulk of a bubble. We suppose the high-frequency noisy component of the electric current (about 100 kHz) seen in Fig.2 is associated with instability of the current filaments.

If neglect noisy current component, one can say that electric breakdown in a single gas bubble forms almost periodical current pulses of a trapezoidal shape although DC applied voltage of a power supply is practically constant or slowly diminishes. Typical period of such breakdown pulsations is rather long –about 0.5 s.

The measurements of the voltage drop across the bubble (taking into account the voltage drop across the liquid) show its increasing with diminishing the current over each period of the pulsations (see Fig.2, left side). One can see -when electric current riches to critical value, it drops down quickly. Opposite, the voltage drop across bubble at this moment increases fast to the breakdown magnitude, and breakdown occurs again.

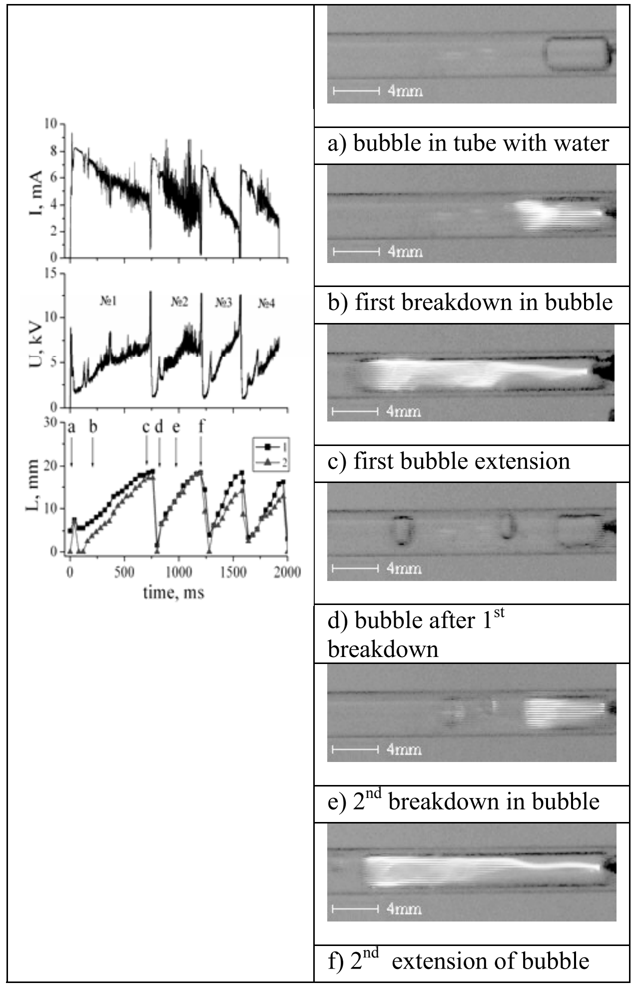

Fig. 2. Electrical and visual characteristics of gas breakdown in a single bubble inserted in dielectric tube with tap water. Applied voltage U_0 is 20 kV. Left side: correlated in time current (I), voltage (U) and lengths (L) of gas bubble (curve 1) and gas discharge column (curve 2) under periodical electric breakdown. Right side: instant pictures of bubble and gas discharge under periodical electric breakdown; the letters a-f correspond to the same letters in left side marking the moments of shots. Exposure time of each shot is 20 ms.

It was found out these pulses are correlated with the periodical extension and shrinking of gas bubble in its length (see Fig.2, left side). In the case of a bubble located near anode, the bubble shrinks to its initial size (in some cases, after breakdown the bubble breaks down to form several small bubbles, Fig.2d, right side). The bubble located in the middle of a tube does not shrink to the initial size. One can see that current averaged over high-frequency noise decreases with an increase in length L of bubble. Opposite, average voltage drop U across bubble grows with increase in length of bubble.

We suppose that volt-ampere characteristic of gas discharge in the extending bubble I=I(U) can be approximated with formula:

$$I = I_0 \frac{U_{max} - U}{U_{max} - U_0}, \qquad (1)$$

where U_0 and U_{max} correspond to the currents $I=I_0$ and $I=0$ respectively.

Indeed, Fig.3 shows that linear dependence between the electric current I and voltage drop U across the bubble is adequate to the experimental data.

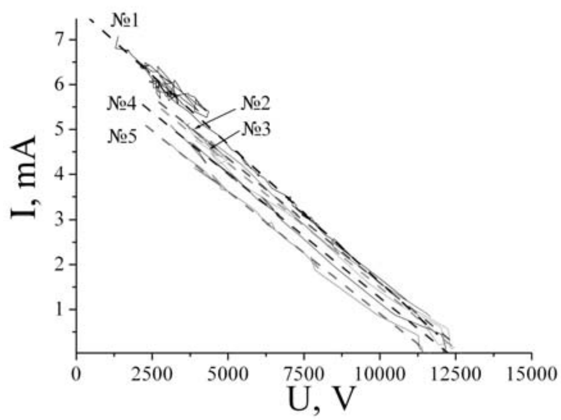

Fig.3. Averaged in time experimental volt-ampere characteristics (solid curves) of gas discharge in bubble over different breakdown periods and their linear approximations (dashed curves). Each number on curve corresponds to the same number of breakdown period marked in Fig.2.

To clarify such behavior of gas discharge in the bubble under each breakdown period, we performed some additional experiments presented in the next partition.

4. Results of the auxiliary experiments

We assume that free-running pulsations of a gas bubble associated with the breakdown and electric discharge inside bubble have the following nature: after electric breakdown, evaporation of a liquid occurs that results in growing gas pressure inside bubble and its expansion; water vapor is the electron attaching gas, therefore the increase in the length of a bubble (i.e. increase in total intensity of the electron attaching processes) results in diminishing the electron number density and current drops down to zero; on the extinction of discharge inside a bubble, water vapor condensates rapidly, the bubble length diminishes to its initial size, and new breakdown in a bubble repeats.

To prove this assumption, we need to estimate a contribution of different parts of a gas discharge (cathode region, plasma column, anode region) in the evaporation of a liquid inside the bubble. Having this in mind, we have done additional experiments with the metallic pin-to-water surface discharge in accordance with the scheme shown in Fig.4.

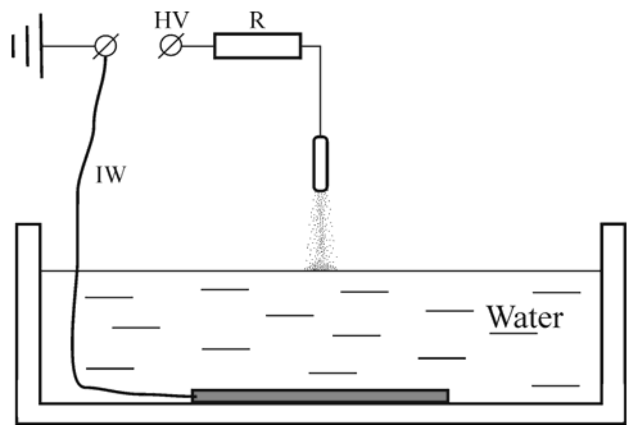

Fig. 4. Scheme of the auxiliary experiments with metallic pin-to-water surface discharge. Liquid used is tap water. IW is the wire covered with high-voltage insulation. R is ballast resistor.

Due to variation of both the inter-electrode distance l between pin and water surface and the polarity of high voltage applied to the pin, we are able to measure the voltage drops across cathode, U_c, and anode, U_a, regions at the water surface (see Fig. 5). The U_c and U_a values were estimated under extrapolation of the discharge voltage to the inter-electrode distance l=0. As the result of these experiments, we found out that $U_c \cong 500$ V and $U_a \cong 100$ V. These figures are agreed with ones published in the literature [11-12].

We measured also the quantity of a liquid (in mg) evaporated during 5 min by the cathode and anode regions separately. It was found out that efficiency of anode region in evaporation of water is equaled to zero but the efficiency of cathode region is equaled to 100%.

We suppose this essential difference of anode and cathode regions in the evaporation of water is associated with different sorts of the current carriers in these regions contacting the water surface: light electrons in anode region and heavy positive ions in cathode region.

Indeed, total intensity of the attachment processes for electrons depends strongly on the reduced electric field strength E/n_g. Within near-electrode layers, the E/n_g magnitude is much higher compared with that in bulk of the plasma. Under high value of the E/n_g, detachment processes predominate over the attachment processes. Due to this, light electrons are the main carriers inside an anode layer. As to glow cathode region, the positive ions are the main current carriers because of low efficiency in the secondary electron emission of the surface bombarded by positive ions.

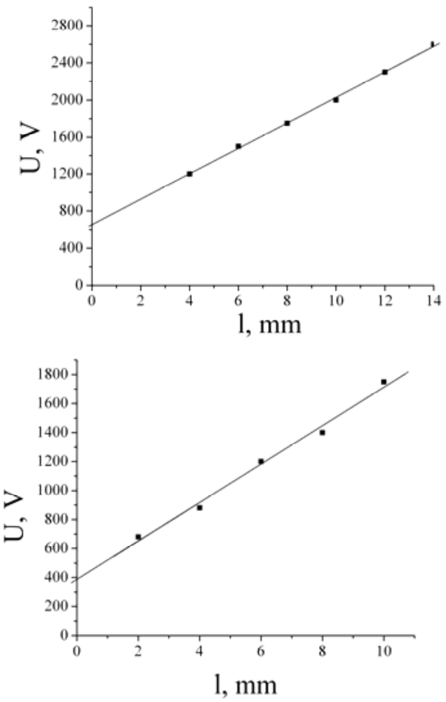

Fig.5. Total voltage of metallic pin-to-water surface discharge at current I=10 mA vs inter-electrode gap. Conditions: tap water, ambient air, room temperature. a), b) corresponds to pin at positive, negative polarity.

5. Discussion

Equations of the model. Because of great (exponentially) dependence of both the ionization and attachment frequencies on the reduced electric field strength E/n_g in gas discharge plasma containing high concentration of water vapor, it is reasonably to assume that magnitude of the E/n_g in a gas discharge is approximately constant (E is the electric field strength, n_g is total number density of the neutral gas inside a bubble). In such a case, it can be shown that voltage drop U across whole length of the plasma column is proportional to total number N of the neutral particles inside the bubble: U ~ N (note, due to changing in a curvature of the bubble poles, the bubble length can be slightly differ from the length of plasma column).

Taking into account a linear dependence between current I and voltage drop U as it was deduced above from the basic experiments (formula (1)), one can write balance equation for total number N of the neutral particles due to the evaporation of a liquid inside the bubble at the cathode region:

$$\chi \frac{dN}{dt} = IU_C = I_0 U_C \left[1 - aN\right] \qquad (2)$$

Here χ is the energy needed to evaporate a single molecule of H_2O, the *a* is the dimensional constant, U_C is the cathode voltage drop, I_0 is the current after breakdown. It is interesting to note that equation (2) does not contain the length of a bubble. As a result, one can obtain the formula for dependence of the current in time:

$$I \cong I_0 \exp(-at) \qquad (3)$$

where $a = I_0 U_c / \chi (N_{max} - N_0)$, N_{max} and N_0 are maximum and initial number of the neutral particles inside bubble.

Assuming that gas pressure inside a bubble is constant (in our opinion, about 3-4 atmosphere) under its extension, we can write the formula (4) for a dependence of the bubble length in time:

$$l(t) \cong l_0 \sqrt{\frac{[N_{max} - (N_{max} - N_0)\exp(-at)]U \exp(-at)}{N_0 U_0}} \qquad (4)$$

Here l_0 is initial length of the bubble.

Comparison between experimental and model results.

A comparison between experiment and simplified theory is presented in Fig. 6. One can see that model results describe not bad the experimental ones. It means the model takes into account really important things like evaporation of a liquid by cathode layer. Note that evaporation of liquid in a bubble is determined by total electric charge transferred through a bubble; therefore high frequency current pulsations have no strong influence on slow behavior of the bubble length in time.

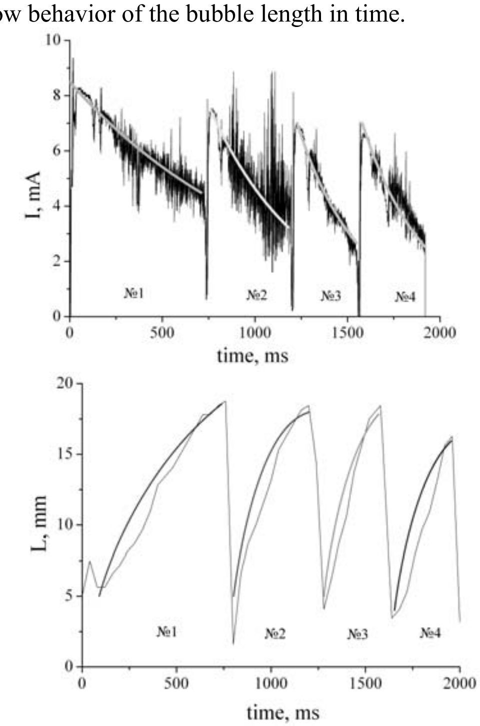

Fig.6. Comparison experimental and modeling results. a) the dependence of an electric current I in time for sequence of repetitive breakdowns inside bubble; solid light curves are numerical results in accordance with formula (3). b) the dependence of the bubble length L in time for sequence of repetitive breakdowns inside bubble; smooth curves are numerical results in accordance with formula (4).

The fitting parameter **a** is the same for the current I (formula (3)) and the length L (formula (4)) at the same gas discharge stages numbered by No 1-4.

6. Conclusions

We have revealed experimentally self-running low-frequency pulsed regime of a DC electric discharge in a single gas bubble inserted in the dielectric tube filled with a conductive liquid. The electric current pulsations are accompanied with the pulsations of the length of gas bubble. The gas bubble length pulsations have the following nature: after each electric breakdown, the evaporation of a liquid occurs that results in the growing gas pressure inside a bubble and its expansion; water vapor is the electron attaching gas, therefore the increase in the length of a bubble (i.e. the increase in total intensity of the electron attaching processes) results in diminishing the current down to zero; on the extinction of discharge inside the bubble, water vapor quickly condensates, the bubble length diminishes to its initial size, and new breakdown repeats again. Such pulsing regime is very attractive and effective from the point of view of intensifying in the generation and hashing of the bio-chemically active species in the liquids.

Acknowledgements

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References

[1] B. Sun, M. Sato, J.S. Clements, Journal of Electrostatics, Elsevier Science Society **39**, 189-202 (1997).
[2] T. Fujii, M. Rea, Vacuum, Pergamon Press **59**,228-235 (2000).
[3] M. A. Malik, A. Ghaffar, S. A. Malik, Plasma source Sci. Technolog, Institute of Physics Publishing. **10**, 82-91 (2001).
[4] A. T. Sugiarto, T. Ohshima, M. Sato, Thin Solid Films, Elsevier Science Society **407,** 174-178 (2002).
[5] M. Monte, F. De Baerdemaeker, C. Leys, A. I. Maximov, Czechoslovak Journal of Physics, Springer **52**, 724-730 (2002).
[6] Masato Kurahashi, Shiniji Katsura and Maizuno. J. Electrostatics, Elsevier Science Society **42**, 93-105, (1997)
[7] Satoshi Ihara, Tomoaki Michi, Saburon Saton, Chobbei Yamabe and Eiji Sakai., Jpn.J.Appl.Phys., **38**, 4601-4604 (1999)
[8] A. M. Anpilov, E. M. Barkhudarov, Yu. B. Bark et al, Journal of Physics, Institute of Physics Publishing **34**, 993-999 (2001).
[9] Yu. S. Akishev, G. I. Aponin, M.E. Grushin et al, Plasma Physics Reports, MAIK Nauka Interperiodica **32**, 12 1052-1061 (2006).
[10] S. Gershman, O. Mozgina, A. Belkind, K. Becker, E. Kunhardt, Contr. Plasma Phys. **47** 1-2 (2007)
[11] F. M. Gaisin, E. E. Son, Yu. I. Shakirov. Volume discharge in vapor-gas media between solid and liquid electrodes. All-Union Polytechnic Institute Publishing House, Moscow, 1990.
[12] A. V. Chlustova, A. I. Maximov, Electronic Treatment of Materials **5**, 35-40 (2002).

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