Influence of diaphragm configuration on DC diaphragm discharge breakdown in electrolyte solution

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Abstract. This paper deals with generation of diaphragm discharge in water solutions of sodium chloride with fixed conductivity of 275 µS. The photos were recorded by high-speed iCCD camera at defined times synchronized by the current passing through the system. Three different phases in the current-voltage curve were recognized under all conditions. Only electrolysis proceeds during the first phase at the lowest applied voltage (300 – 1200 V depending on the orifice dimensions), while bubbles were created thanks to the intense Joule heating inside the orifice during the second phase. Finally, the discharge was ignited at applied voltages over 1600 V. These facts were confirmed by iCCD images taken during all three phases. We concluded, by comparing current-voltage characteristics of different orifice diameter sizes, that this parameter had an important influence on the bubbles generation phase. On the contrary, the diaphragm thickness played an important role at the discharge breakdown.

1. Introduction
Pin hole discharge configuration consists of two electrode spaces divided by the dielectric barrier with a central pin-hole. Mechanisms of discharge breakdown in liquids are still under an intensive research and their study requires specific approach in all possible configurations (different kind of high voltage, various electrode configuration, etc.). Generally, two types of theories are considered to be the most suitable for discharge breakdown description – thermal (bubble) theory, and electron theory [1].

Discharge in the pin hole configuration probably combines both theories. Initially, it starts in the pin-hole when sufficient power is applied. The breakdown moment is probably related to the bubbles formation [2]. By the application of constant DC voltage, water solution is significantly heated due to high current density (Joule’s heating), and microbubbles of water vapor are created in the pin-hole region. It is assumed that the discharge breakdown starts inside these bubbles because of high potential gradient between the outer and inner bubble region [2], in correspondence to the thermal theory. On the other hand, further propagation of plasma channels to the bulk solution probably corresponds to the electron theory. Moreover, application of DC voltage initiates creation of two kinds of plasma streamers on both sides of the dielectric barrier [3][4].

2. Experimental setup
The batch plasma reactors using a diaphragm configuration with total volume of 100 ml are employed in the presented work. The discharge was created in an orifice (a pin-hole) in the dielectric barrier
separating two electrode parts of the reactor. DC non-pulsed high voltage up to 2 kV was used for the discharge generation. Electrodes were made of stainless steel and they were installed in parallel to the diaphragm from the dielectric barrier in each reactor part. The dielectric barrier was Shapal-MTM − ceramics made with the variable thickness (0.3-2 mm). One pin hole at the diaphragm centre with diameter of 0.3-1 mm was used in experiments. Water solution containing NaCl electrolyt to provide initial conductivity of 275 $\mu$S·cm$^{-1}$ was used as a liquid medium.

Time resolved characteristics of current, voltage and images camera were recorded using four-channel oscilloscope which detected their output values. Oscilloscope LeCroy LT374L (4-channels) operating up to 500 MHz with high voltage probe ELDITEST GE 3830 30kV, 1:1000 was used to obtain resolved characteristics of discharge voltage. Another probe 1:1 was used for current measurement using ballast resistor with resistance of 5.13 $\Omega$. Simplified scheme of oscilloscope integration into the electrical circuit is demonstrated in figure 1. Mean values of breakdown parameters (voltage, current, power and resistance) were calculated and subsequently, static current-voltage characteristics were constructed for each experiment. We also used ICCD camera iStar, with program Andor iStar and objective Cosmicar/Pentax TV Lens 50 mm, 1:2,8, for the discharge imaging; camera synchronization was based on current passing through the system. We set either the positive or negative peak, which give signal over the Pulse Generator TTi, 10 MHz TGP 110 to the camera. The camera captures with records over 0.3 ms.

Figure 1. Simplified scheme of electrical circuit containing discharge reactor and oscilloscope: Electrical scheme of the experiment: 1 – discharge reactor, 2 – dielectric barrier with pin-hole, 3 – anode, 4 – cathode, 5 – oscilloscope LeCroy LT374L, F – fuse (0.5 A), C – microwave capacitor (0.85 $\mu$F), R – resistances: $R_3$ (100 M$\Omega$), $R_4$ (3.114 k$\Omega$), $R_5$ (5.13 $\Omega$), $R_6$ (105.5 $\Omega$), $R_7$ (0.13 $\Omega$).

3. Results and discussion
Current-voltage characteristics of DC pin hole discharge were constructed from the mean values of time resolved current and voltage records over 10 ms. Figure 2 demonstrates a typical current-voltage curve obtained in NaCl solution with initial conductivity of 270 $\mu$S·cm$^{-1}$. This curve could be divided into following three parts. Initially at low applied voltages, increasing the applied DC voltage, measured current increased slightly and more or less directly proportionally. The time resolved current record depicted on figure 3 shows relatively smooth line and thus we can conclude that only electrolysis took place in the system. Of course, the electrolyte solution was heated by the Joules effect due to the passing current. Going above the voltage of some hundreds volts, the first significant breakpoint appeared in the curve – current gently jumped up. According to the time resolved characteristics and photo demonstrated in the figure 4(a), the smooth current time record changed and some current pulses can be recognized. These current pulses were related to the substantial creation of micro bubbles formed by the evaporating solution inside the pin hole where the current density was the highest and thus the Joule heating was sufficient for microbubble creation as shown in the photo. Further enhancement of voltage provided only a small current increase until the second remarkable breakpoint was observed. From this moment, current was rapidly arising with only a small voltage enhancement. This second breakpoint was assigned to the discharge breakdown moment and with further voltage increase discharge started to operate more regularly.
Figure 2. Typical current-voltage characteristic of DC diaphragm discharge in NaCl solution with initial conductivity of 275 µS·cm⁻¹ and 0.7 mm barrier thickness with pin-hole diameter of 0.3 mm.

Figure 3. Time resolved current record in NaCl solution with mean voltage values of 860 V.

Figure 4. Time resolved characteristic of voltage and current with photo in NaCl solution: mean voltage values of (a) 1530 V; (b) – (d) 1700 V

Time resolved characteristics and photos for different discharge phases are demonstrate in figures 4 (b) – 4 (d). Figure 4 (b) demonstrates random discharge breakdown with light emission at the current peak maximum as it is shown on the photo. Figure 4 c) demonstrates bubbles with ignited discharge created around the pin-hole when current reached its maximum. The photo in figure 4 (d) demonstrates that current decrease is followed by weakening of light emission and subsequent bubble expansion.
As it is shown in figure 5 in phase of electrolysis the current through the pin-hole is higher with the pin-hole diameter increasing at constant voltage and diaphragm thickness. Start bubbles formation were slightly increased from 700 V for diameter of 0.3 mm to 900 V for diameter 0.6 mm and 1000 V for diameter 1 mm. Breakdown voltage as well as current were increased with increasing the pin-hole diameter. The measured breakdown voltage for 0.3 mm pin-hole diameter is 1200 V; for diameter 0.6 mm is 1300 V; for diameter 1.0 mm is 1400 V.

The increase of the barrier thickness has a substantial effect only on the discharge breakdown part of the current-voltage curve as it is shown in figure 6. During electrolysis and bubble formation phase the increasing of the barrier thickness has a negligible effect on the current. However, the breakdown voltage increases from 1300 V (at thickness of 0.3 mm) to about 1500 V for 0.7 mm thickness and to about 1800 V for 1.5 mm thickness.

![Figure 5. Comparison of current-voltage characteristics of the diaphragm discharge in NaCl solutions with conductivity of 275 µS·cm⁻¹ for three pin-hole diameters diaphragm.](image)

![Figure 6. Comparison of current-voltage characteristics of the diaphragm discharge in NaCl solutions with conductivity of 275 µS·cm⁻¹ for three different barrier thicknesses.](image)

**4. Conclusion**

Breakdown moment, as well as processes of electrolysis and bubbles formation were identified from obtained statistic and time resolved characteristics of DC diaphragm discharge and time resolved photos. Current-voltage evaluation was remarkably influenced by using different pin-hole diameter and barrier thickness. Increase of diaphragm diameter has a major effect on the electrolysis. Increase of diaphragm thickness determines discharge breakdown as well as its operation.

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**References**

[1] Malik M A, Ghaffar A, Malik S A 2001 *Plasma Sources Sci. Technol.* **10** 82.
[2] Joshi R P, Qian J, Schienbach K H 2002 *J. Appl. Phys.* **92** 6245
[3] Stará Z, Krcma F, Procházková J 2008 *Acta Technica CSAV* **53** 277.
[4] Hlavatá L, Krcma F, Kozáková Z 2011 *Book of Contributed Papers: 18th Symposium on Application of Plasma Processes and Workshop on Plasmas as a Planetary Atmospheres Mimics*, Vrátma 2011, pp. 83–87 (id 18960)