

PIN HOLE DISCHARGE CREATION IN Na₂SO₄ WATER SOLUTIONS

LUCIE HLAVATÁ^a, RODICA SERBANESCU^{a,b}, LENKA HLOCHOVÁ^a,
ZDENKA KOZÁKOVÁ^a, FRANTIŠEK KRČMA^{a,*}

^a Brno University of Technology, Faculty of Chemistry, Purkyňova 118, 612 00 Brno, Czech Republic

^b Faculty of Physics, Ovidius University of Constanța, 124, Mamaia Boulevard, 900527 Constanța, Romania

* corresponding author: krcma@fch.vutbr.cz

ABSTRACT. This work deals with the diaphragm discharge generated in water solutions containing Na₂SO₄ as a supporting electrolyte. The solution conductivity was varied in the range of 270 ÷ 750 μS cm⁻¹. The batch plasma reactor with volume of 100 ml was divided into two electrode spaces by the Shapal-MTM ceramics dielectric barrier with a pin-hole (diameter of 0.6 mm). Three variable barrier thicknesses (0.3; 0.7 and 1.5 mm) and non-pulsed DC voltage up to 2 kV were used for the discharge creation. Each of the current–voltage characteristic can be divided into three parts: electrolysis, bubble formation and discharge operation. The experimental results showed that the discharge ignition moment in the pin-hole was significantly dependent on the dielectric diaphragm thickness. Breakdown voltage increases with the increase of the dielectric barrier thickness.

KEYWORDS: pin hole discharge, discharge in liquids, discharge breakdown.

1. INTRODUCTION

Electrical discharges in liquids have been in a serious focus of researchers for mainly last three decades. Especially formation of various reactive species such as hydroxyl and hydrogen radicals, some ions, and molecules with high oxidation potential (hydrogen peroxide) has been investigated in order to utilize this process in water treatment, removal of organic compounds from water, and sterilization processes [8, 9, 5, 4].

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 [3].

Discharge ignition 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 [1]. 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 [1], 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 [7, 2].

Longer streamers appear on the side with the cathode because the pin-hole in the dielectric barrier represents a positive pole (like point in point to plane configuration), and the streamers propagate to the positive electrode similarly to the positive corona discharge. On the other side where the anode is placed, shorter streamers in a spherical shape propagate towards the pin-hole like in the case of the negative corona discharge [6].

The presented paper describes the pin hole discharge creation by means of electrical characteristics, and discusses the influence of the dielectric barrier thickness on the breakdown moment and current voltage characteristic.

2. EXPERIMENTAL SET-UP

The batch reactor was divided into two electrode spaces by the dielectric barrier, and non-pulsed DC voltage up to 2 kV was used for the discharge creation. The discharge appeared in a pin-hole in the dielectric diaphragm. The dielectric barrier was made of Shapal-MTM ceramics with the three different thicknesses (0.3; 0.7 and 1.5 mm). The pin-hole diameter was 0.3 mm and it remained constant during the whole experiment. Planar electrodes (40 × 30 mm) made of stainless steel were installed on each side of the barrier. Water solution containing Na₂SO₄ electrolyte to provide initial conductivity in the range of 270 ÷ 750 μS cm⁻¹ was used as a liquid medium. Total volume of the used solution was 100 milliliters (50 milliliters in each electrode space). Solution temperature was changed by the discharge operation,

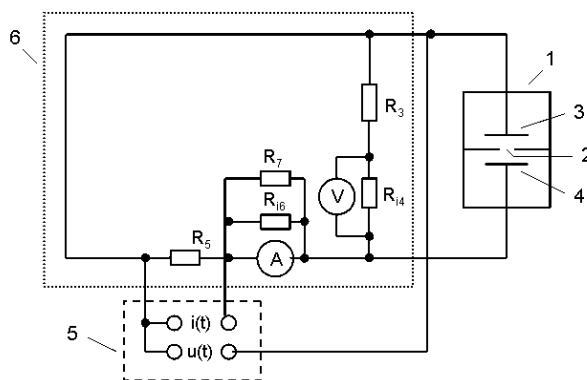


FIGURE 1. Electrical scheme of the experiment: 1 – discharge reactor, 2 – dielectric barrier with pin-hole, 3 – anode, 4 – cathode, 5 – oscilloscope Tektronix TDS 1012B, 6 – DC HV source with resistances important for electric measurements: R_3 ($100\text{ M}\Omega$), R_{i4} ($3.114\text{ k}\Omega$), R_5 ($5.13\ \Omega$), R_{i6} ($105.5\ \Omega$), R_7 ($0.13\ \Omega$).

but its enhancement was up to 10 K, only, during each measurement. This temperature change was negligible in term of the discharge breakdown.

Oscilloscope Tektronix TDS 1012B operating up to 100 MHz with Tektronix P6015A high voltage probe was used to obtain time resolved characteristics of discharge voltage and current with focus on the breakdown moment. The scheme of the electric circuit including diagnostics is demonstrated in Fig. 1. Mean values of breakdown parameters (voltage, current, power, and resistance) were calculated and subsequently, static current–voltage characteristics were constructed for each experiment. The obtained results were compared with respect to the electrode configuration (barrier thickness) and to the electrolyte conductivity.

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 50 ms. Figure 2 demonstrates a typical current–voltage curve obtained in Na_2SO_4 solution with initial conductivity of $550\ \mu\text{S cm}^{-1}$. This curve could be divided into following three parts.

- (1.) Initially at low applied voltages, increasing the applied DC voltage, measured current increased more or less directly proportionally. The time resolved current record shows smooth line and thus we can conclude that only electrolysis took place in the system. Of course, due to the passing current the electrolyte solution was heated by the Joule effect.
- (2.) Going above the voltage of some hundreds volts, the first significant breakpoint appeared in the curve – current markedly jumped up. According to the time resolved characteristics, the smooth

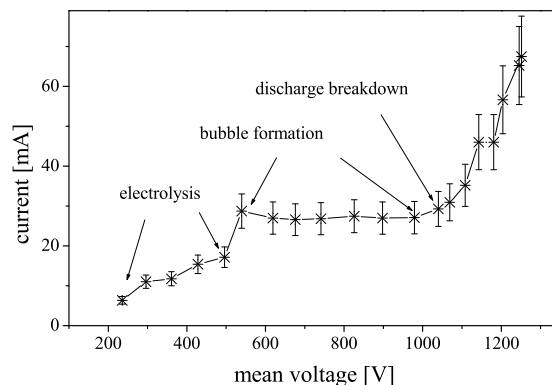


FIGURE 2. Typical current–voltage characteristic of DC diaphragm discharge in Na_2SO_4 solution with initial conductivity of $550\ \mu\text{S cm}^{-1}$ and 0.3 mm barrier thickness.

current time record changed and some current pulses can be recognized. We have assumed that 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. Further increase of voltage provided only a small current increase because the bubbles creation started to be more or less regularly until the second significant breakpoint was observed.

- (3.) From this moment, current was rapidly arising with only a small voltage increase. This second breakpoint was assumed to be the discharge breakdown moment which was also confirmed by light emission recorded by the optical emission spectroscopy.

Mean values over 50 ms of voltage and current were estimated from time resolved characteristics and subsequently, current–voltage curves were constructed. Figure 3 demonstrates the comparison of current–voltage characteristics obtained for three different barrier thicknesses (0.3; 0.7 and 1.5 mm) in electrolyte solutions (Na_2SO_4) at conductivity of $270\ \mu\text{S cm}^{-1}$.

The increase of the barrier thickness had a substantial effect on all three parts of the current–voltage curve. Curves obtained with the barrier thickness bigger than 0.3 mm were located at the lower current values. The reason could be explained by the increase of resistance with the increasing barrier thickness. The presented current–voltage curves show that the discharge breakdown voltage was enhanced by the increasing of the barrier thickness. The particular values of determined breakdown voltage are listed in Tab. 1 (in the second column) for three barrier thicknesses (0.3; 0.7 and 1.5 mm). Breakdown voltage increased from 1000 V (thickness of 0.3 mm) to about 1500 V for 1.5 mm thickness. These results

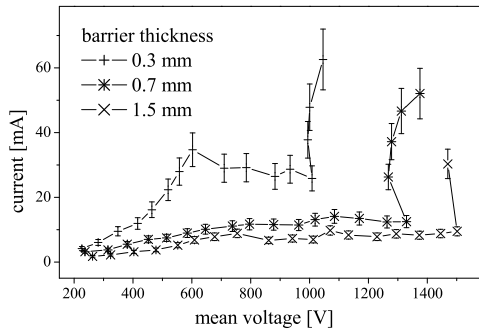


FIGURE 3. Comparison of current–voltage characteristics of the pin hole discharge in Na_2SO_4 solutions (conductivity of $270 \mu\text{S cm}^{-1}$) for three different barrier thicknesses.

barrier thickness [mm]	conductivity [$\mu\text{S cm}^{-1}$]		
	270	550	750
0.3	990 V	1040 V	1020 V
0.7	1270 V	1130 V	1200 V
1.5	1470 V	1370 V	1290 V

TABLE 1. Breakdown voltage of DC pin hole discharge as a function of the barrier thickness for the selected conductivities.

were obtained in Na_2SO_4 solution (initial conductivity of $270 \mu\text{S cm}^{-1}$). Similar effect was observed for the other conductivities (550 and $750 \mu\text{S cm}^{-1}$) using the same electrolyte solution, too. The determined breakdown voltages for these conductivities are listed in Tab. 1 (in last two columns).

Figure 4 demonstrates the time evaluation of voltage and current at the mean voltage of 1000 V for three different barrier thicknesses. These figures clearly demonstrate the difference between the diaphragm (thickness of 0.3 mm, ratio l/d 0.5) and capillary discharge (thickness of 1.5 mm, $l/d = 2.5$). Resistance inside the pin-hole increased and current reached lower values with the increasing barrier thickness. Time resolved characteristic for 1.5 mm thickness (Fig. 4c) shows the electrolysis, only. It is well visible that there are no significant peaks appearing in regular voltage and current oscillations. Voltage oscillations were related to the HV source construction, and they had no significant influence on the observed phenomena.

Remarkable higher oscillations of both current and voltage were recorded at the barrier thickness of 0.7 mm (Fig. 4b). The current record shows nearly regular shape of the oscillations without any significant current peak. This phenomenon was related to the micro bubble formation due to the intensive solution heating by passing current. No light emission was observed during this period.

The irregular shape of current peaks with some high current peaks appeared if the thinnest barrier

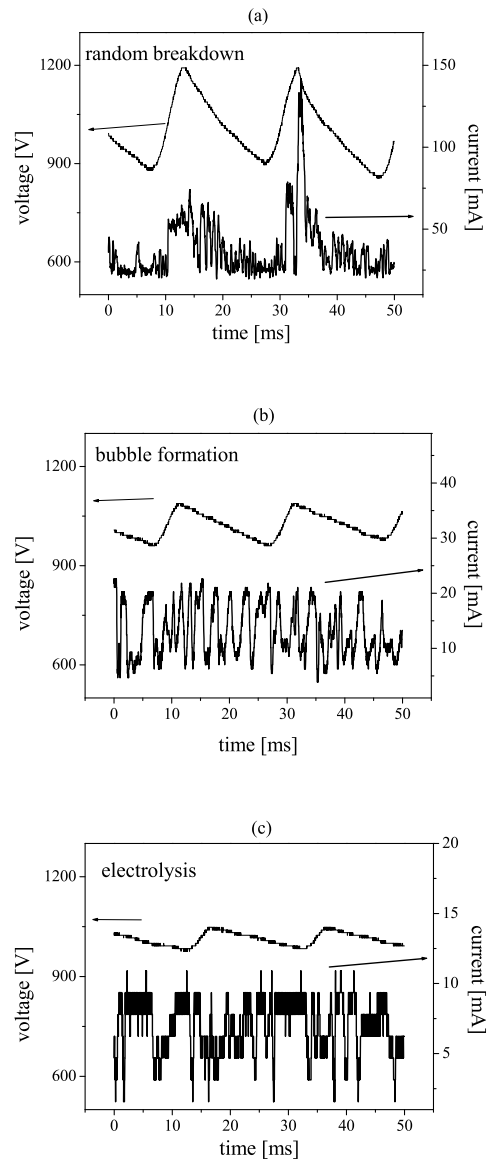


FIGURE 4. Time resolved voltage and current records for mean voltage of 1000 V in Na_2SO_4 solutions (initial conductivity of $270 \mu\text{S cm}^{-1}$) using the dielectric barrier with thickness of a) 0.3 mm, b) 0.7 mm and c) 1.5 mm.

was applied (Fig. 4a). This phenomenon was related to the random discharge breakdown in vapor bubbles. Simultaneously, the short peaks of emitted light were recorded, too. As the pin hole diameter was greater than the barrier thickness, the pin hole represented a significantly lower resistance. Therefore, current in the pin hole was much higher which effect led to an easier discharge ignition in the pin hole.

4. CONCLUSIONS

Breakdown moment as well as processes of electrolysis, and bubble formation were identified from obtained mean value current voltage characteristics; the time resolved characteristics clarified the proposed mech-

anisms of the pin hole discharge creation in Na₂SO₄ solutions. Current–voltage evaluation was remarkably influenced by the dielectric barrier configuration. Breakdown voltage increased with the increase of the dielectric barrier thickness. Current–voltage curves were shifted to the lower currents and higher voltages with the increase of the barrier thickness. This effect was caused by the increase of resistance with the increasing barrier thickness. Solution conductivity had only a minor effect on the discharge characteristics. Thus we can conclude that the pin hole geometry is the main parameter influencing the bubbles formation as well as the pin hole discharge breakdown.

ACKNOWLEDGEMENTS

This work was supported by the Czech Ministry of Culture, project No. DF11P01OVV004.

REFERENCES

- [1] R. P. Joshi, J. Qian, K. H. Schoenbach. Electrical network-based time-dependent model of electrical breakdown in water. *J Appl Phys* **92**(10):6245–6251, 2002.
- [2] Z. Kozakova, L. Hlavata, F. Krcma. Diagnostics of electrical discharges in electrolytes: Influence of electrode and diaphragm configuration. In *Book of Contributed Papers: 18th Symposium on Application of Plasma Processes and Workshop on Plasmas as a Planetary Atmospheres Mimics*, pp. 83–87. Vrátna, 2011.
- [3] M. A. Malik, A. Ghaffar, A. M. Salman. Water purification by electrical discharges. *Plasma Sources Sci Technol* **10**(1):90–97, 2001.
- [4] M. Moisan, J. Barbeau, M.-C. Crevier, et al. Plasma sterilization. Methods and mechanisms. *Pure Appl Chem* **74**(3):349–358, 2002.
- [5] E. Njatawidjaja, A. T. Sigianto, T. Ohshima, M. Sato. Decoloration of electrostatically atomized organic dye by the pulsed streamer corona discharge. *J Electrostat* **63**(5):353–359, 2005.
- [6] J. Prochazkova, Z. Stara, F. Krcma. Optical emission spectroscopy of diaphragm discharge in water solutions. *Czech J Phys* **56**(2 supplement):B1314–B1319, 2006.
- [7] Z. Stara, F. Krcma, J. Prochazkova. Physical aspects of diaphragm discharge creation using constant DC high voltage in electrolyte solution. *Acta Technica CSAV* **53**(3):277–286, 2008.
- [8] B. Sun, M. Sato, J. S. Clements. Oxidative processes occurring when pulsed high voltage discharges degrade phenol in aqueous solution. *Environ Sci Technol* **34**(3):509–513, 2000.
- [9] P. Sunka, V. Babicky, M. Clupek, et al. Potential applications of pulse electrical discharges in water. *Acta Phys Slovaca* **54**(2):135–145, 2004.