Acta Technica CSAV 53 (2008), 277–286

PHYSICAL ASPECTS OF DIAPHRAGM DISCHARGE CREATION USING CONSTANT DC HIGH VOLTAGE IN ELECTROLYTE SOLUTION[1]

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Abstract. New results were obtained from the investigation of the breakdown moment of diaphragm discharge generated using constant DC voltage in water solutions of two electrolytes (NaCl and $NaNO_3$). Electrical discharge was created in an orifice (initial diameter of 0.2 mm) in dielectric diaphragm (thickness of 0.25 mm) separating two electrode spaces. Both dynamic and static VA characteristics were recorded and, subsequently, breakdown parameters were determined as a function of solution conductivity (adjusted by electrolyte concentration in the range of 300–1300 $\mu S\,cm^{-1}$). Obtained results revealed a significant decrease of breakdown voltage (from 1350 to 880 V) and resistance with the increasing solution conductivity. On the other hand, discharge power and current at breakdown moment was enhanced by the increasing conductivity (from 40 to 100 mA). This effect was similar in both tested electrolytes. Discharge ignition in water was related with bubble creation in the orifice, according to the thermal theory of electrical discharge generation. This phenomenon was confirmed by records of high speed camera and sound diagnostics.

Key words. Diaphragm discharge in electrolyte solutions, electrical characteristics of breakdown moment.

Introduction

Electrical discharges generated in liquids require different initial conditions for their breakdown than electrical discharges in gases. This difference is mainly caused by higher liquid density (and thus particle concentration) and dipole momentum of water molecules (if the discharge is created in water solutions). But there are more factors acting in the discharge initiation. Due to this fact, limited electrode configurations are convenient for discharge generation

[1]This research has been supported by the Czech Science Foundation, projects No. 202/07/P371 and No. 202/03/H162.

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using DC voltage in water solutions. The most frequent configurations investigated are point-to-plate or coaxial geometry [1]–[3], capillary discharge [4] or diaphragm discharge [5]. Majority of scientists have studied these discharges generated using DC pulsed voltage. Thus our work has been focused on the initiation of diaphragm discharge by constant DC voltage.

This paper describes breakdown moment in so-called diaphragm discharge which is created in a small pin-hole in the dielectric barrier separating two electrode spaces. As DC high voltage is applied on the electrodes, two kinds of plasma channels (streamers) propagate from the pin-hole (i.e. the discharge initiation spot) towards electrodes, each on the opposite side of the diaphragm. For better understanding of streamer propagation, there is an analogy between the generation of the diaphragm discharge and corona discharge in point-to-plate electrode configuration. The pin-hole behaves as a point electrode of both polarities each one for each part of the reactor. In the part with the plane cathode, electrons are accelerated towards the positively charged pin-hole (like to the point anode) and the remaining positive space charge at the end of the previous electron avalanche further enhances the electron velocity. Thus created streamers represent long plasma channels and we call this part "positive discharge". On the other side, electrons propagating from the negatively charged pin-hole (like from the point cathode) towards the plane anode are dragged by the positive space charge remaining on the end of the previous electron avalanche and thus the electron velocity decreases. The final shape of negative streamers is represented by shorter plasma channels in a spherical shape and we called it "negative discharge". Moreover, positively charged ions in the system are attracted by the cathode but their propagation velocity is much slower than the velocity of electrons. A simplified scheme of plasma channels propagating on both sides of the diaphragm is given in Fig. 1. Plasma streamers generated in this way differ in their shape, electron velocity as well as in the energy dissipation [6]. Therefore processes consequently initiated in both electrode spaces are strongly influenced by this streamer disparity.

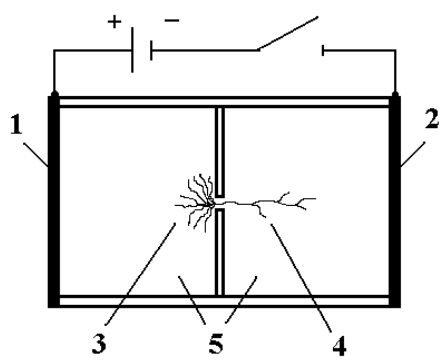

Fig. 1. Simplified scheme of DC diaphragm discharge; 1—anode, 2—cathode, 3—negative plasma channels, 4—positive plasma channels, 5—conductive liquid

Initiation mechanisms of diaphragm discharge breakdown in water solutions can be describe by two types of theories: thermal (bubble) and electron theory [7]. The first theory is based on the fact that liquid is intensively heated (by Joule heating) in the region of high electric field that is concentrated in the pin-hole in the diaphragm. When water evaporates breakdown appears in evaporated bubbles due to the substantial voltage gradient on bubble surface (interface between gas and liquid phase). Subsequently, discharge is propagated in liquid phase by electron avalanches like according to Townsend's theory for electrical discharge in gases. This paper confirms validity of both theories using records made by high speed camera and photograph with high resolution.

The first results focused on static VA characteristics of diaphragm discharge breakdown in electrolyte solutions have been already published in our previous work [8]. This paper extends our study by dynamic characteristics in two electrolyte solutions (NaCl and $NaNO_3$) and describes the breakdown moment in more details (from the viewpoint of electrical parameters, sound and optical diagnostics). Moreover, influence of solution properties on discharge breakdown and propagation is presented.

Experimental

A simple batch discharge reactor constructed according to the scheme in Fig. 1 was used in our experiments. Its total volume of 3.5 litres was divided into two parts by the dielectric diaphragm made of chemically inert as well as sufficiently hard and elastic material (PET, thickness of 0.25 mm) with a small pin-hole in the centre (initial diameter of 0.2 mm). Two planar high voltage electrodes made of stainless steel were installed symmetrically on both sides of the diaphragm in the distance of 2 cm from the diaphragm. The discharge was created in the pin-hole of the diaphragm using the DC high voltage source which gave the constant voltage up to 2 kV. No cooling and mixing system was used.

Tested solutions were prepared by dissolving of selected electrolyte (NaCl, $NaNO_3$) in distilled water. Initial solution conductivity was adjusted by appropriate electrolyte concentration in the range of 300–1300 $\mu S\,cm^{-1}$. Solution pH was not specially modified and its initial value was approximately neutral.

Electrical measurements were carried out by two channel digital storage oscilloscope Tektronix TDS 1012B operating at 100 MHz ($1\,GS\,s^{-1}$). High voltage probe Tektronix P6015A was used for records of discharge voltage. Dynamic characteristics of discharge voltage and current were recorded with the focus on the breakdown moment. Mean values of breakdown parameters (voltage, current, power and resistance) were calculated using specially created program and subsequently, VA characteristics were determined for each experiment. Moreover, sound loggings were recorded by the same device using the microphone instead of the HV probe. Optical diagnostics providing images of the discharge formation was carried out by high speed digital camera.

Results and discussion

It has been already presented that ignition of electrical discharge (particularly diaphragm discharge) in water solutions using constant DC voltage was highly dependent on solution conductivity [9]. Low conductivity adjusted by low concentration of electrolyte induced relatively high resistance of the system and disabled the discharge breakdown. On the other hand, too high conductivity values caused such low resistance that it was also insufficient for the breakdown appearance. Based on these facts, series of dynamic characteristics were recorded around the breakdown point in solutions of two electrolytes (NaCl and $NaNO_3$) with initial conductivity varying from 300 to 1300 $\mu S\,cm^{-1}$. Consequently, influence of this parameter on the breakdown characteristics was observed. Each experimental series started at zero voltage input from the source and it was progressively enhanced by approximately 50 V until the breakdown appeared. Both voltage and current time evaluation in the reactor were recorded at each step and mean values of these parameters as well as power and resistance were also calculated for each step.

Figure 2 clearly demonstrates the breakdown appearance in the NaCl solution (initial conductivity of 700 $\mu S\,cm^{-1}$). Increasing input voltage from HV source, both voltage and current in the reactor also increased. However, current enhancement went much slowly than the increase of voltage. When the input voltage exceeded the breakdown moment, current started to increase rapidly. On the other hand, discharge voltage oscillated around some mean value which further remained more or less constant.

Looking on the breakdown point in details, significant difference in current evaluation can be seen (Figs. 3–5). The first record of this series (Fig. 3) represented the moment just before the breakdown appearance in NaCl solution (initial conductivity of 700 $\mu S\,cm^{-1}$). Mean values of voltage and current in the reactor were 1030 V and 72 mA, respectively. When the first attempts of breakdown appeared in the bubbles in the pin-hole remarkable peaks of current were observed together with the decrease of system resistance and related drop of voltage (Fig. 4). Mean value of voltage did not differ from the previous step a lot (1040 V) but the mean value of current significantly increased to 116 mA. Time evaluations of voltage and current during the regular discharge propagation are demonstrated in Fig. 5. Both increase of current and decrease of voltage and resistance were determined simultaneously. Mean value of voltage slightly decreased to 1013 V but current increased to 170 mA. Significant peaks of current were related to the discharge formation in bubbles of evaporated solution (more details are discussed below).

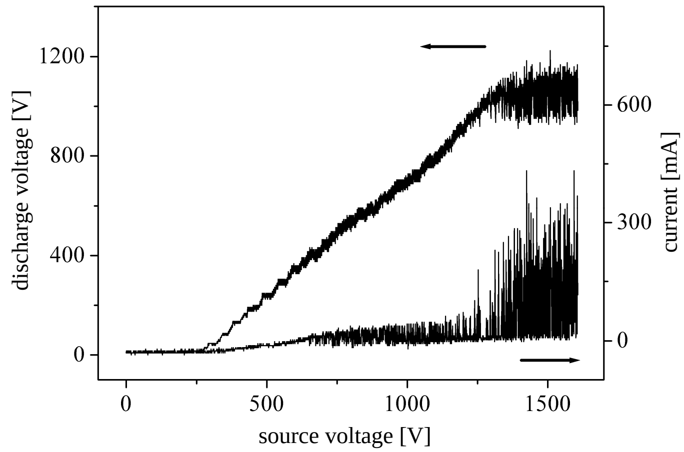

Fig. 2. Discharge voltage and current as a function of increasing input voltage from the source (NaCl solution, initial conductivity of 700 μS cm^{-1})

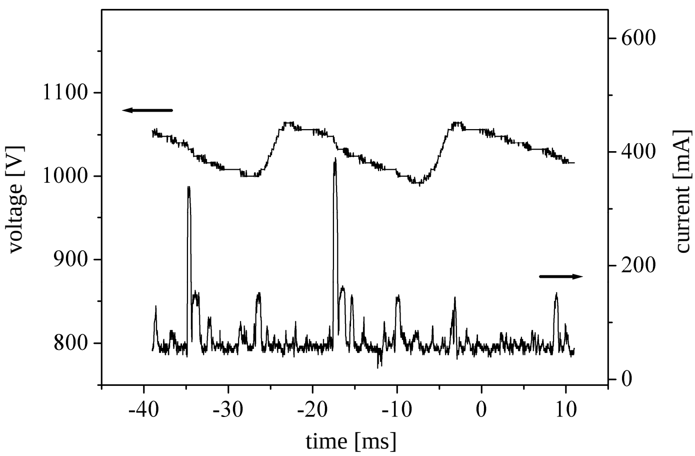

Fig. 3. Dynamic characteristic of voltage and current in the diaphragm reactor (NaCl solution, initial conductivity of 700 μS cm^{-1}, mean values: 1030 V, 72 mA)

It has been already mentioned in the introduction that there are two theories describing the initiation of electrical discharge in liquids—thermal (bubble) and electron theory. To confirm the first theory, e.g. bubble creation and discharge ignition in water vapour, special medial experiments focused on image and sound diagnostics were carried out in our reactor. Records of high speed camera revealed the bubble formation in the pin-hole as well as the different shape of streamers propagating on both sides of the diaphragm [10]. These images confirmed the thermal theory of discharge creation: water solution was

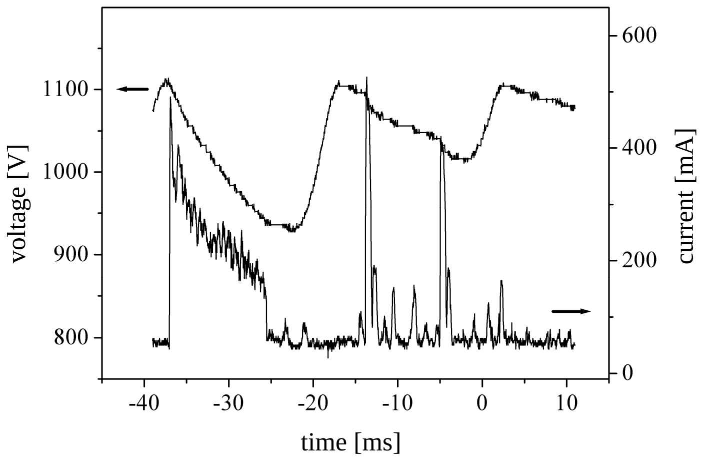

Fig. 4. Dynamic characteristic of voltage and current in the diaphragm reactor (NaCl solution, initial conductivity of 700 μS cm^{-1}, mean values: 1040 V, 116 mA)

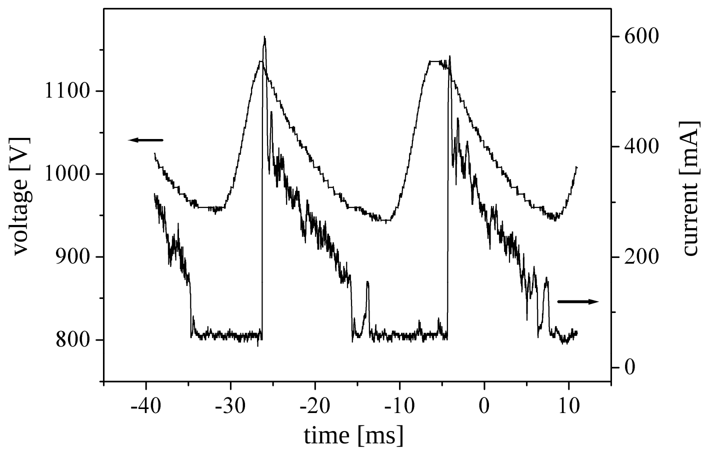

Fig. 5. Dynamic characteristic of voltage and current in the diaphragm reactor (NaCl solution, initial conductivity of 700 μS cm^{-1}, mean values: 1020 V, 170 mA)

intensively heated by the applied electric field, bubbles of evaporated solution formed in the pin-hole vicinity (the spot with the highest electric intensity) and breakdown primary appeared in these bubbles. As bubble lifetime was short (it expanded), discharge also turned off with bubble splitting but it was subsequently ignited in another bubble. Thus the diaphragm discharge regime showed a specific quasi-pulsed character although it was generated using constant DC voltage. Moreover, we had come to the idea that breakdown of the discharge in bubble could be closely related to a noise response. We carried out a special experiment with sound registration and results of obtained records are presented in Fig. 6 together with current evaluation. It is evident that current peaks (corresponding to the breakdown in the bubble) were in quite a good accordance with sound pulses. Therefore we can assume that the thermal theory of discharge ignition in bubbles was suitable for our discharge.

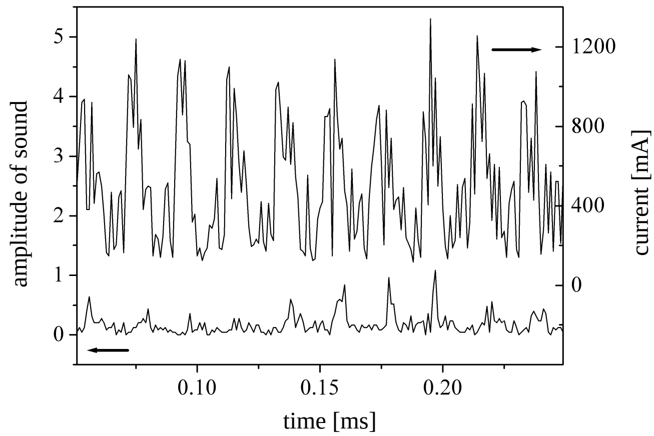

Fig. 6. Sound and current records of the diaphragm discharge in NaCl solution (700 µS cm^{-1}); sound data are in absolute values

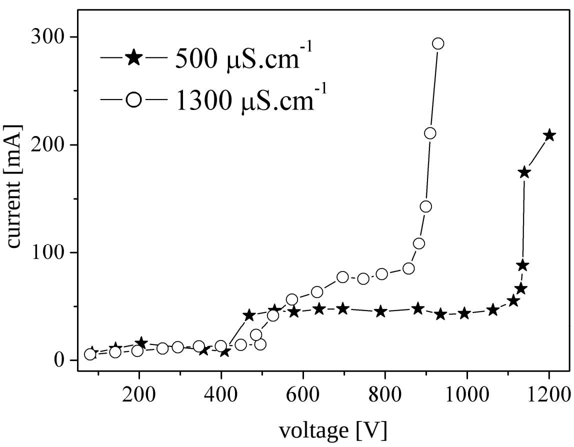

Fig. 7. VA characteristics of diaphragm discharge in NaCl solution of two conductivities (500 and 1300 µS cm^{-1})

Static VA characteristics were obtained from the calculation of mean values (voltage, current, power and resistance) from the dynamic measurements.

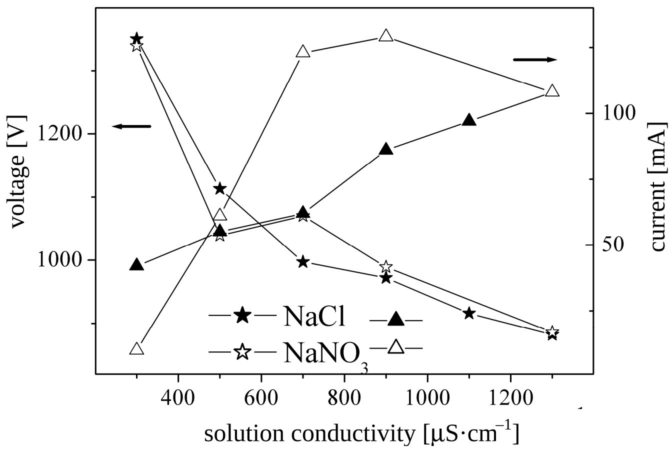

Fig. 8. Breakdown mean values of voltage (star) and current (triangle) as a function of initial conductivity of NaCl (full symbols) and $NaNO_3$ (open symbols) solutions

Determination of breakdown moment was realized for each VA curve of all tested solutions. Series of two electrolytes (NaCl and $NaNO_3$) with initial conductivity from 300 to 1300 $\mu S\,cm^{-1}$ were investigated and compared. An example of typical VA characteristics in NaCl solution of two selected conductivities (500 and 1300 $\mu S\,cm^{-1}$) is given in Fig. 7. Significant break in the curve determines the moment of discharge ignition. Comparing two solution

Table 1. Breakdown parameters of DC diaphragm discharge in NaCl solution of various initial conductivities (mean values of voltage, current, power and resistance in the reactor)

Conductivity [$\mu S\,cm^{-1}$]	Voltage [V]	Current [mA]	Power [W]	Resistance [kΩ]
300	1350	42	54	38.0
500	1113	55	61	24.8
700	997	62	67	16.2
900	972	86	87	13.4
1100	916	97	90	11.7
1300	882	108	96	9.7

conductivities in Fig. 7 it is clearly shown that this moment started at different voltage and current magnitude. Obtained breakdown parameters for NaCl solutions of various conductivities are summarised in Table 1. It is evident that discharge ignition in solutions with higher initial conductivity started at lower voltage than in low conductive solutions. On the other hand, breakdown current slightly increased with the increasing conductivity. This effect was related to lower resistance of liquid in the case of higher conductive medium and vice versa. Breakdown power behaved similarly as current so its value was enhanced by increasing solution conductivity. Comparing VA characteristics of the discharge in both selected electrolytes, similar results were obtained in $NaNO_3$ solution like in NaCl. Breakdown voltage and resistance were decreasing with the increasing initial conductivity, following more or less the same tendency. However, breakdown current and power dependence on conductivity revealed a particular difference in the enhancement caused by the increase of this parameter. Comparison of breakdown voltage and current for both electrolytes is given in Fig. 8. It is evident that breakdown current was increasing almost linearly with the increasing conductivity of NaCl solution. In $NaNO_3$ solution current enhancement by conductivity was more intensive until approximately 600 $\mu S\,cm^{-1}$. Over this conductivity value, breakdown current stopped to increase or even became to decrease. This ascertained phenomenon is quite new and it will be an object of our further research focused on the breakdown in another electrolyte kinds.

Summary and conclusions

Diaphragm discharge was generated using constant DC voltage up to 2 kV. Its ignition was observed in the orifice (initial diameter of 0.2 mm) in dielectric barrier separating two electrode spaces. Breakdown phenomenon was studied in two electrolytes (NaCl and $NaNO_3$) with initial conductivity varied from 300 to 1300 $\mu S\,cm^{-1}$. Both static and dynamic electrical characteristics were evaluated as well as image and sound records to describe breakdown moment in the solution.

Results obtained from high speed camera, digital photography and microphone records confirm the thermal theory of discharge generation in liquids, e.g. initial breakdown started in bubbles of water vapour. Moreover, images declared formation of different plasma streamers propagating on both sides of the dielectric diaphragm.

Dynamic characteristics clearly showed the evaluation of electric parameters (especially voltage and current) around the breakdown point. Increasing the input voltage from the source, both voltage and current constantly increase, too. Getting over the breakdown moment, current started to increase rapidly in significant pulses while voltage remained more or less constant oscillating around some mean magnitude.

Influence of solution conductivity on breakdown appearance was observed. Breakdown parameters (voltage, current, power and resistance) were determined from VA characteristic of each experiment. While both breakdown voltage and resistance were decreased by the increasing solution conductivity, increase of conductivity enhanced breakdown current and power. Similar results were obtained in both tested electrolytes however, current dependence on conductivity revealed a slight difference between these two electrolytes and this effect will be an object of our further research.

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[Received September 22, 2008]