Nonthermal plasma in contact with liquids. Electrical breakdown.

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Abstract: Low temperature plasmas are extensively used in materials production, environmental and biomedical applications. Nanosecond discharges allow generation of nonthermal plasma without substantial water heating and are promising in variety of applications where operation temperature is naturally limited (biomedicine) or in order to reduce the operation costs (water treatment). We present a study of nanosecond discharges in underwater point-to-plane configuration. The discharge is characterized by electrical measurements, shadowgraphy, ICCD imaging and time resolved spectroscopy.

Keywords: discharges in water, nanosecond discharge, shadowgraphy

1. Introduction

Mechanisms of underwater discharge initiation and propagation are of a great interest in pulsed systems, water treatment and biomedical applications. Nanosecond discharges allow generation of nonthermal plasma without substantial water heating. For some applications high repetition rate is required provoking interest in discharge breakdown in the gaseous channel structures remaining from the precedent voltage pulse. Dielectric recovery of distilled water was studied for repetition rates up to 4 KHz [1], however, for pulses delay exceeding several microseconds filamentary discharge pattern is almost turned into the multitude of bubbles. Discharges in dielectric liquids were extensively studied for microsecond voltage pulses. Different morphologies of positive and negative streamers have been reported [2]. However, the mechanisms of underwater streamer initiation are still disputable. Commonly, two mechanisms were considered: a micro or even nanobubble formation by Joule heating and direct electron impact of liquid, however, in submicrosecond region impact ionization in pre-existing microbubbles assumed to be the most likely explanation [3]. Recently it has been reported [4] on observation of sub-nanosecond anode-initiated plasma discharge which propagation velocity was found to be 5000 km/s over first 200 ps after ignition. Such rapid discharge development could not be ascribed to water vaporization and, in author’s opinion, is relevant to impact ionization of liquid.

2. Experimental setup

The experimental setup allows up to 2 ns time resolution of shadowgraphic imaging and emission spectroscopy in the range between 200 and 900 nm. The discharge device is designed in order to fit the limited volume of 10 mm quartz cell (Hellma) and to minimize electrical noise from the discharge.

Discharge is performed in the quartz cell filled with a 3 ml of distilled water (50 $\mu$S/cm). Discharge configuration consists of nickel plated pin electrode with a tip diameter of 65 $\mu$m soldered to the core of a 50 Ohm high voltage (HV) cable (RG213). Grounded electrode is a U-bent copper stripe of a 5 mm width connected to the HV cable shield. Discharge device is immersing in the water till all the inter-electrode volume is completely filled with the liquid and all residual air bubbles are avoided. Grounded electrode is oriented in such a way that its plane is parallel to the windows of the cell providing direct optical access to the discharge. The distance between HV tip and semicircular part of U-like grounded electrode is about 3 mm.

HV pulses of negative polarity with 20 kV of amplitude, 5 ns rise time and 30 ns duration (FWHM) (FID FPG10–1MKS20) are applied with a frequency in the range 1-100 Hz. In the case of a positive tip negative pulse is applied to the U-bent copper stripe and grounded cable shield is connected to the pin electrode. Two back current shunts (BCS) consisted each of 13 low-inductance resistance of 2.2 Ohm are installed in the HV cable.
The first BCS is placed 1 m after the nanosecond pulse generator and the second is inserted in the middle of the 25 m long cable. Cable length of 25 m results in approximately 250 ns time separation between first generated pulse of negative polarity and consecutive twice-reflected positive pulse. As water gap resistance is infinite compared to cable impedance of 50 Ohm, the pulse amplitude is doubled when pulse reaches the water gap. Hence, we obtain a series of 3 HV pulses of -20, +18 and -16 kV peak voltage on the high voltage electrode with 250 ns of separation between them. Energy dissipated in plasma during each of three reflected pulses is calculated from BCS signals. Typical shunt signal obtained with LeCroy (iX64) oscilloscope is presented on fig.1.

![Figure 1](image.png)

**Figure 1.** Typical back current shunt signal. I, II and III are three successive HV pulses coming to the needle electrode (see detailed description in the text).

The fast shadowgraphic imaging is performed using an ANDOR iStar DH734 ICDD camera allowing the minimum acquisition gate of 2 ns. The xenon light sources (PerkinElmer PAX-10) is used as a backlight and a parallel light beam is formed by doublet lens with a pinhole. Quartz cell with a submerged discharge device is irradiated by a parallel beam and a 6 cm quartz lens is then forming a magnified shadow image of a discharge on the photocathode of the ICDD camera. Time resolved emission spectra are measured by ANDOR Shamrock SR-303i spectrometer with mounted ANDOR iStar DH734 ICDD camera as a detector in the spectral range 190-900 nm (1800 l/mm grating). Due to the low luminosity of the discharge averaging over 500 acquisitions is necessary in order to obtain an acceptable signal/noise ratio.

In the imaging mode two pulse generators (TTi TGP 10) are used to synchronize HV pulse generator with a xenon lamp with a pulse duration of 2 μs. Flash lamp pulse is controlled with a fast photodiode. The signal from the first BCS is used to trigger the camera gate. Time synchronization is ensured by visualization of the photodiode signal, BSC signals and iCDD camera gate monitor with the oscilloscope.

### 2. Results of experiments

![Figure 2](image.png)

**Figure 2.** Typical shadowgraph images for a bush-like discharge mode. a) 5 ns, b) 15 ns and 40 ns after discharge ignition. Time resolution is 2 ns.

In the case of first negative pulse we observe two possible structures on the discharge shadowgraphs:
bush-like discharge type with a typical size of several tens of micrometers and tree-like filamentary discharge pattern exceeding 1 mm in size. Typical images are given by figs.2 and 3.

Both discharge types demonstrate a supersonic propagation speed with a pronounced exponential decrease during the applied voltage pulse. Filamentary mode develops with a speed of about 50 km/s (see fig.4) at early ignition stage and almost stops to propagate after the HV pulse end. Filamentary gas channels observed in our experimental conditions are of the order of 5 micrometers in diameter. They are enveloped in a series of circular compression waves with a typical step of 50 micrometers between two successive waves. These experimentally deduced parameters of fast filamentary mode demonstrate its scale difference from underwater filamentary streamers observed by other authors for microsecond discharges [2] typically for applied voltage pulses in microsecond time scale.

Bush-like discharge type is slower compared to the filamentary mode and has a maximum propagation velocity 4 km/s at the stage of the discharge initiation (fig. 5). Propagation of this discharge mode, similar to a fast filamentary mode, stops after HV pulse end. At the late discharge development
phase a single or a pair of circular pressure waves with a center at the discharge ignition point on the HV tip can be seen. The speed of these compression waves is found to be 1.55 km/s (fig. 5) which is of the order of sound velocity in the water (1.45 km/s).

Statistical investigation shows that both discharge types are equiprobable and their probabilities do not depend on the repetition rate of applied voltage in the range 5-0.1 Hz claiming that residual bubbles from the previous shot do not influence the ignition. The probabilities of each discharge mode neither change with a time which means that accumulation of plasma discharge products does not at the origin of one of the two modes. However, the transition between two modes may occur and is observed during the trailing edge of first negative HV voltage pulse, when a filamentary structure emerges from the bush-like discharge type. Statistically the probability of such a discharge mode transition is less than 1%. Energy dissipated in plasma is calculated from the second BCS signal. Tree-like mode is characterized by 1.9 mJ of adsorption energy for the first negative pulse which is 0.3 mJ higher than energy dissipated in bush-like discharge.

Tree-like mode is characterized by strong light emission during the leading and trailing edge of the voltage pulse which prevents shadowgraphic analysis at the very first stage of ignition. At the contrary bush-like discharge is weakly luminescent. Spectroscopic study demonstrates characteristic intense features of N$_2$ (2$^+$ and 1$^-$) and weak OH emission. Analysis of emission pattern shows a bright spot just above the HV tip and less luminous filamentary structure. N$_2$ bubble formed from the dissolved nitrogen may be at the origin of strong emission and its size variation from on shot to another could be responsible for different discharge modes.

Anode-initiated discharge (a positive tip) represents only one typical discharge shape: two hemispherical structures of the order of several micrometers in diameter. This discharge is also characterized by spherical compression waves emission and like negative bush-like discharge is weakly luminescent.

The successive negative pulse can give rise to bush-like and tree-like modes.

4. Conclusions

Successive discharges of alternating polarities in distilled water were studied. Cathode-initiated discharges develop in two possible configurations: slow and weakly luminescent bush-like mode or fast and luminous tree-like mode with well-pronounced branching. Under our experimental conditions, the two modes originate with approximately equal probability. Positive discharge demonstrates only one discharge morphology with two hemispherical weakly luminescent structures. Successive negative pulse results in formation of one of negative modes, bush- or tree-like mode.

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References


