

# Plasmas in liquids and at the interfaces

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## Introduction

Historically the early research on electrical breakdown of liquids followed two distinct purposes: the prevention of sparking in liquid isolators and production of thermal plasma for underwater welding and generation of strong shock waves. Experimental studies were mainly focused on formation of conductive channel in the discharge gap filled with liquid, while the initiation and prebreakdown phenomena were almost inaccessible with the experimental facilities of that time [1]. Later, extensive experimental study performed in wide range of liquid dielectrics, including pure liquid hydrocarbons, mineral oils, water and liquefied gases at cryogenic temperatures demonstrated the existence of two distinct discharge propagation structures: slower bushy mode and faster filamentary mode, both consisted of gaseous channels [2 and ref. herein]. For positive discharge with the typical rising time of applied HV of 10 ns it was shown that slow mode systematically originated at lower voltages as compared to the fast mode. Moreover the maximal propagation velocity of the fast mode was found to be insensitive to the hydrostatic pressure up to the  $10^6$  Pa whereas the velocity of slow mode decreased drastically [3]. For longer HV rising time (lower dV/dt) fast filamentary mode could only ignited after the formation of slow mode [4]. So it was suggested, that propagation dynamics of slow mode was governed by hydrodynamic expansion of the discharge cavity assisted by EHD forces acting on the charged interface [5]. For the fast mode field induced dissociation and electron impact ionization of bulk liquid were proposed [6 and ref. herein]. However Monte Carlo calculations [7] demonstrated that the electron multiplication cannot occur under the fields typical for the fast mode initiation in water without assuming a local density lowering. Recent numerical study [8] demonstrated that for polar liquids in highly nonuniform fields with high dE/dt rate typical for nanosecond pulses, the strong electrostriction effect induces pressure gradient and eventually can lead to the rupture of liquid in the electrode vicinity. Such mechanism can contribute at the discharge initiation stage and can provide sufficient local density decrease at the head of propagating fast mode channel. This work aims at investigation of the very first stage of the discharge initiation in liquid dielectric under nanosecond HV pulse.

## Experimental setup

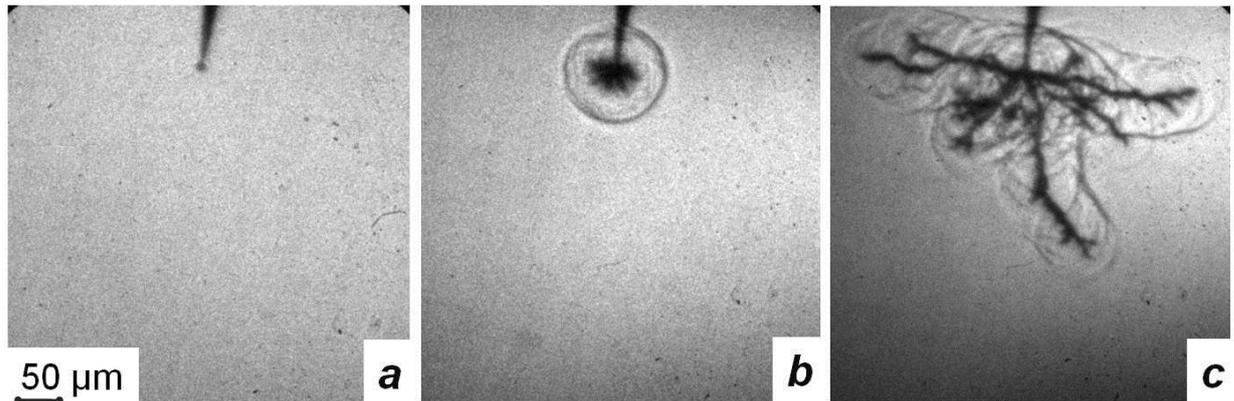
Time-resolved shadowgraphic and optical emission visualization was applied to study the prebreakdown phenomena occurring on the point electrode in point-to-wire geometry under high voltage nanosecond pulse. The exhaustive description of experimental setup can be found elsewhere [9]. Briefly, high voltage pulses of 30 ns duration with 5 ns rising slope and 1.5 – 9 kV amplitude were applied on  $\varnothing 2$   $\mu\text{m}$  nickel tip. Due to the impedance mismatch at the discharge gap the voltage is doubled on the tip. Andor iStar DH734 iCDD camera with 2 ns time resolution was synchronized with a HV pulse giving overall jitter better than 1 ns. Three liquids with different dielectric constants were chosen for the test: deionized water ( $\epsilon=80$ ), ethanol

## Results and discussions

Three different scenarios can be observed in polar deionized water ( $\epsilon = 80$ ) and ethanol ( $\epsilon = 27$ ) depending on the applied voltage: i) formation of gaseous spherical cavity, ii) initiation of slower bush-like and iii) faster tree-like discharge on the point anode.

It is shown that for the voltage amplitude in the range of 3 – 4kV in deionized water and 3.5 – 4.5 kV in ethanol, a spherical gaseous cavity with a radius of about 4  $\mu\text{m}$  is formed on the electrode tip (Figure 1a) after the end of the applied voltage pulse. For the lowest voltage values cavity expansion dynamics can be approximated by Rayleigh model. Deduced initial pressure in the cavity is found to be 3 MPa in water and 1 MPa in ethanol. In n-pentane ( $\epsilon = 1.8$ ) a spherical

cavity of 5  $\mu\text{m}$  was detected after the HV pulse of 10 – 12 kV. Experimentally observed cavitation in deionized water, ethanol and n-pentane under the nanosecond high voltage pulse with maximal electric field rate of  $0.6 - 2,6 \cdot 10^9 \text{ V/m}\cdot\text{ns}$  (3 – 12 kV, 5 ns rise time) is in good agreement with the recent theoretical predictions made by Shneider with coworkers [8]. Electrostrictive negative pressure stretching the liquid in the direction of the electric field gradient can exceed the cavitation threshold for given liquid dielectric. For three tested liquids the product  $\epsilon E^2$  which determines the maximal electrostrictive pressure is proportional to the cavitation threshold values available in literature [10]. Although, no light emission or shock wave formation was observed in the case of bubble cavitation, other mechanisms including evaporation due to the Joule heating and field induced Zener process [6] cannot be completely discarded.



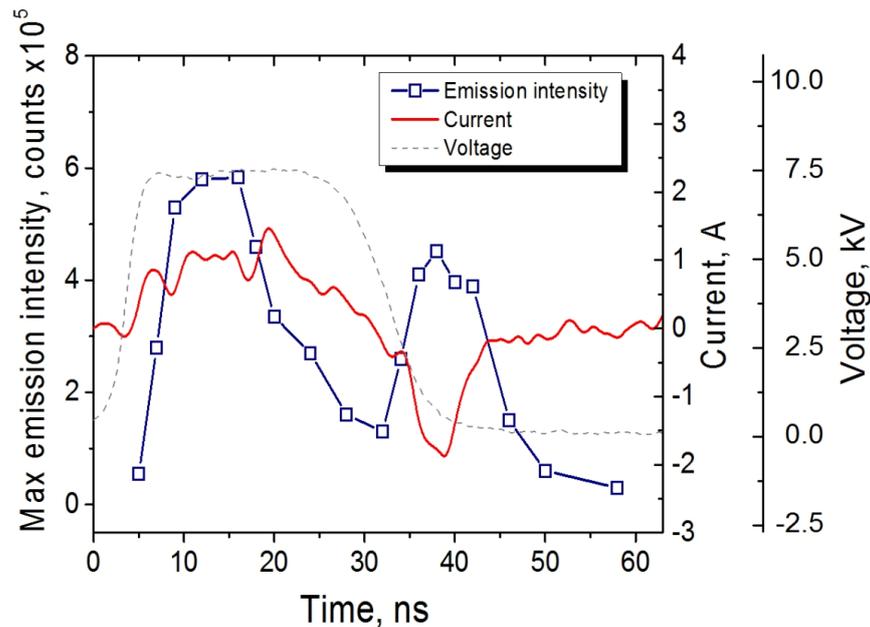
**Figure 1:** a) Cavitation mode at 4 kV b) Slow bush-like mode at 9 kV c) Fast tree-like mode at 9 kV. Deionized water

At higher applied voltage of 4 kV – 5.5 kV a bush-like discharge can be observed in deionized water (Figure 1b) and ethanol. At the initial phase of the bush-like discharge development the propagation velocity is supersonic. The maximal velocity at ignition increases with the applied voltage from 2.2 km/s at 4 kV reaching 4.2 km/s at 9 kV in deionized water and from 2 km/s at 5.5 kV up to the 3 km/s at 9 kV in ethanol. Discharge propagation is accompanied by emission of a spherical shockwave. Initial discharge pressure in deionized water estimated from the maximal shock wave velocity gives 0.8 GPa and 5.7 GPa at 4 kV and 9 kV respectively. High initial pressure indicates that a strong energy release occurs at the discharge initiation. Field induced dissociation and autoionization of liquid molecules in the vicinity of the tip can be responsible for the formation of the bush-like discharge. The density lowering due to the electrostrictive force can provide local strengthening of reduced field promoting electron avalanche formation.

The bush-like discharge consists of a gaseous cavity and few filaments radiating from the cavity. The total structure is quite similar in deionized water and ethanol. The number of discharge filaments and their maximal length increase with the applied voltage. Discharge stops to propagate at the end of plateau of the applied voltage pulse. Collapse of channel extremities can be observed at low applied voltage of 4 kV. Maximal excessive pressure inside the bush-like discharge filaments at the stopping moment can be estimated from the surface tension and is found to be about 30 kPa. We conclude that the propagation of bush-like discharge is driven by the expansion of the initially formed gaseous cavity, whereas the channel formation can be explained by the interface instability development in highly nonuniform electric field. Bush-like discharge is not observed in n-pentane.

At voltage amplitude higher than 5.5 kV a fast filamentary tree-like discharge can be initiated on the point electrode. Statistically the probability to observe the fast mode increases with the applied voltage between 6 and 15 kV. Light emission associated with the ignition of the tree-like

discharge is detected during the rising slope of the applied voltage pulse (Figure 2) before the shadow structures become visible.



**Figure 2:** Evolution of the plasma emission intensity of the tree-like mode and discharge current at applied voltage of 7.5 kV

The maximal discharge velocity is of the order of 20 – 50 km/s in water and an order of magnitude slower (3 – 5 km/s) in ethanol at 6 – 9 kV. Tree-like discharge is also observed in n-pentane at the voltage amplitude higher than 12 kV. The maximal propagation velocity in n-pentane varies between 2.2 and 4.6 km/s for voltage amplitude in the range of 12 – 18 kV.

The number and length of the discharge channels increase gradually with the applied voltage in all tested liquids. In deionized water energy dissipation in the tree-like discharge at 15 kV is about 0.2 mJ. The maximal discharge current is 1.5 A and the average charge transferred by each channel equals to 2 nC. Time evolution of the light emission from the discharge channels correlates with the total discharge current (Figure 2). A back discharge is observed over the trailing edge of the high voltage pulse as supported by negative current spike and reillumination of discharge channels.

Propagation of the fast tree-like discharge is accompanied by formation of a series of the shock waves along the channel pass. The estimation of pressure in the discharge channel is done based on the channel expansion dynamics and from the shock wave velocity. Two models give consistent values of the maximal discharge pressure of 0.3- 0.4 GPa at 6 kV. The maximal discharge pressure is found to increase drastically with the applied HV amplitude reaching 6 GPa at 15 kV.

Although the electrostrictive effect may facilitate the initiation of tree-like mode it does not seem capable of explaining the difference in the propagation velocity observed in deionized water, ethanol and n-pentane. Especially because under the electric fields at which the tree-like mode is ignited the dielectric permittivity is considerably lowered due to the saturated polarization [11]. The latter effect can improve the nonuniformity and induce the local increase of the electric field. Field induced impact dissociation of liquid molecules in extremely high fields of the order of  $10^9$  V/m is believed to be responsible for the propagation of the tree-like mode. The strong coupling between hydrogen bonding and dipole orientation in the case of water can explain the higher propagation velocity as compared to ethanol. Electric field of the order of  $10^9$  V/m induces strengthening of the hydrogen bonds along the field axis while hydrogen bonds

orthogonal to the field are weakened [12]. Proton tunneling through the hydrogen bonds, known as Grotthuss mechanism, can contribute in the development of the fast discharge mode.

Obtained results demonstrate that depending on amplitude of applied nanosecond voltage pulse three different scenarios can be observed in polar dielectrics. Initiation of nanosecond discharge in liquid dielectrics is a complex phenomenon in which the electro-hydrodynamic and atomic mechanisms are strongly coupled. Hence the models taking into account hydrodynamic and electronic processes have to be developed in the future.

It is found that fast (tree-like) mode can be ignited occasionally during successive reflected pulse of the positive polarity coming about 520 ns after the initial HV pulse while it was never observed for the reflected pulse of the negative polarity. The polarity effect similar to the gas phase streamers [13] is due to the difference in mobility of the electrons and ions. A net negative charge is formed on the interface screening the applied field in the case of negative discharge.

We attempted to apply the optical emission spectroscopy (OES) in order to get insight into the plasma parameters. Spatially and time integrated over the HV pulse duration emission spectra demonstrate a continuum emission between 300 and 800 nm and strongly broadened atomic H $\alpha$  (656 nm) and OI (777 nm) lines. Time evolution of hydrogen alpha profile measured in the successive reflected pulses is consistent with the typical time of pressure relaxation inside the discharge channels. During the first (positive) pulse the H $\alpha$  line had asymmetric red wing and can be approximated with two shifted Lorentzian profiles. Two Lorentzian functions are ascribed to the direct and the back discharge inside the channels of the tree-like structure. Each profile is assumed to be a sum of combined action of the Stark and the Van der Waals broadening. Based on this assumption we obtained an electron density of  $1.3 \cdot 10^{26} \text{ m}^{-3}$  for the direct discharge and  $2 \cdot 10^{25} \text{ m}^{-3}$  for the back discharge.

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