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Non-equilibrium plasma in liquid phase: time-resolved diagnostics and leader-type model based on electrostriction mechanism

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Abstract: In this paper we are focused on studying of the nanosecond-pulsed discharge in liquids with different polarization properties. Schlieren method was used in order to demonstrate formation of negative pressure region in liquids with different dielectric permittivity constants: water, ethanol and ethanol-water mixture. It is shown that this density perturbation, formed at the raising edge of the high voltage pulse, is followed by a generation of a shock wave propagating with the speed of sound away from the electrode, with negative pressure behind it.

Keywords: Schlieren, nanosecond, water, electrostriction, shock waves

1. Introduction

Plasma in liquids has become an attractive field of study over the last decade, particularly for the wide range of potential applications such discharges can have – see for instance the applications discussed in [1]. Paramount to exploring the full potential these discharges can offer is the proper understanding of the mechanisms involved with its inception and development. As research continues in this area, it is becoming more evident that a universal description simply cannot be applied, and that the physical mechanisms related to discharges produced in liquids depend on the experimental conditions under consideration. Discharges ignited with voltage pulses having microsecond or longer rise times to breakdown voltage are preceded by the formation of a low density (bubble) zone near the electrodes. These low-density zones facilitate the reduced electric field conditions necessary to initiate breakdown [2, 3]. When the voltage rise time is much faster – as with nanosecond and sub-nanosecond pulses – plasma is initiated directly in the liquid medium without bubbles [4, 5]. Still, these two cases are only relevant to discharges produced by submerged electrodes, and one needs to bear in mind that other methods for generating plasma in liquids exists, such as laser heating and focused sound waves [1].

In this work we focus our attention on nanosecond pulsed discharges generated between submerged electrodes. One attractive quality of this regime is the minimal temperature increase of the surrounding liquid which ensues as a result of these discharges. Estimates from [4] indicate that the liquid temperature change (locally) from a single pulse is on the order of 50K. The increases the controllability of the conditions under which these discharges can be utilized. Furthermore, the lack of formation of bubbles makes it particularly interesting for applications where gas release might not be desired.

This study seeks to explore the mechanism via which these discharges are initiated. Close observation of the structure of the discharge propagating through the liquid reveals a ‘lightning-like’ appearance – see for instance an ICCD image of a nanosecond pulsed discharge formed in polymer oil (PDMS) in Figure 1**Error! Reference source not found.**. This propagation stage has been explained by gas phase plasma theory – the plasma propagates via a leader mechanism where the filament propagates due to the high electric field in the streamer head [6]. The most intriguing aspect of this work comes from studying the phase that precedes propagation that initiated the discharge. Bubble growth has been experimentally and theoretically ruled out in [4, 5], and the minimum electric field conditions at which discharge is initiated falls well below the theoretically predicted values for condensed media [5].

One theory seeking to explain this phenomenon is the electrostriction mechanism [7-9]. This theory hypothesizes the formation of a region saturated with “nanopores” in the vicinity of the HV electrode tip; these nanopores form as a result of electrostrictive forces acting on the liquid due to the strong, non-uniform electric field. Fast voltage rise times are required to ensure that hydrodynamic forces do not counteract the region of negative pressure within which these voids are formed. The primary purpose of this work is to investigate the plausibility of this mechanism through experimental verification. The authors in [7] predict that at voltages just below breakdown, the electrostriction conditions will be met, and can lead to subtle changes in the isotropicity of the liquid's refractive index. Optical techniques sensitive enough to detect minute changes in refractive index will lend support to this theorem. Thus, we employ Schlieren and shadowgraph techniques to investigate the liquid behaviour in the proximity of the electrode when a nanosecond high-voltage pulse is applied to it. Liquids of varying dielectric strengths are investigated since the model also predicts a direct dependence on the relative permittivity on the liquid.

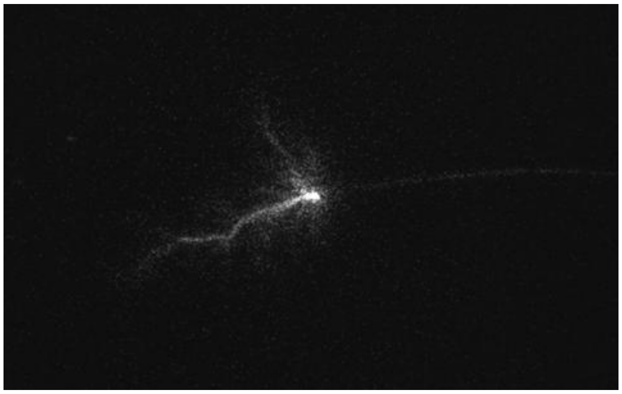

Figure 1: Nanosecond pulsed discharge in PDMS showing lightning-like structure of plasma propagating in the liquid.

2. Experimental Setup

The experimental setup is similar to the one described in [5]. A pin-to-plane electrode configuration was used: the high voltage pin was a mechanically sharpened tungsten rod with a radius of curvature at the tip of 35µm, fixed 3mm away from a grounded copper plate. The chamber contained ~50ml of liquid when filled, and 50mm of liquid layer separated the discharge gap from the quartz viewing windows fitted to the chamber. Two liquids were used in these experiments: deionized water, and ethyl alcohol. A 1:1 volumetric mixture was also used. The effective relative permittivity of each liquid was: 80 (water), 25 (alcohol) and 55(1:1 mix).

Pulses with 8.6kV amplitude (17.2kV due to signal reflection) were delivered to the electrodes via 15m of RG393/U high voltage coaxial cable. Pulses had 4ns rise time, 10ns duration and 5ns fall time, with a maximum pulse repetition frequency of 1Hz used throughout all the experiments. A calibrated back current was used for signal monitoring and synchronization.

Schlieren and shadowgraph experiments were enabled by a 32mW laser diode, operating at 532nm, and the 4-Picos camera (Stanford Computer Optics) served as the high speed detector. The principle schematic of the experimental system is shown in Figure 2. This allowed visualization of density perturbations in the x-y plane to be imaged. These results in these experiments were obtained for an uncalibrated setup, and thus qualitative estimates of the density changes and angles of refraction could not be made.

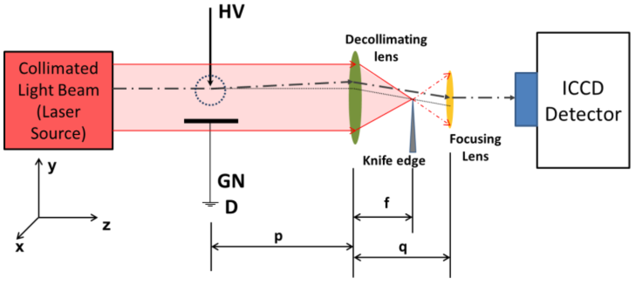

Figure 2: Basic principle of the schlieren transmission imaging experiment.

Under these experimental conditions, no discharge was observed; the electric field was slightly below the threshold required to ignite the plasma in the liquids used. Image sequences with 10ns camera exposure were taken to trace the changes in the liquid's optical density over time.

According to the electrostriction model, the total pressure acting on the region near the electrode as a result of this fast rising high voltage pulse is given by:

$$ - \qquad (1) $$

Here, is the applied electric field, is the liquid dielectric constant, is the density of the liquid. Within the first few nanoseconds of applying the pulse, hydrostatic forces do not have time to counteract the electrostrictive forces, and thus a region of negative pressure develops near the electrode. Our experimental setup was constructed to identify this region of negative pressure with the corresponding compression wave in the region that follows its formation.

3. Results

The results of the experiments performed with pure water are shown in Figure 3 following. The time annotation in the pictures refer to the time relative to application of the voltage pulse, with being the time when the rising edge of the pulse just gets to the electrodes. We see the formation of a density perturbation forming very close to the tip upon the application of the pulse at , which then propagates away from the tip in the form of a compression wave travelling at .

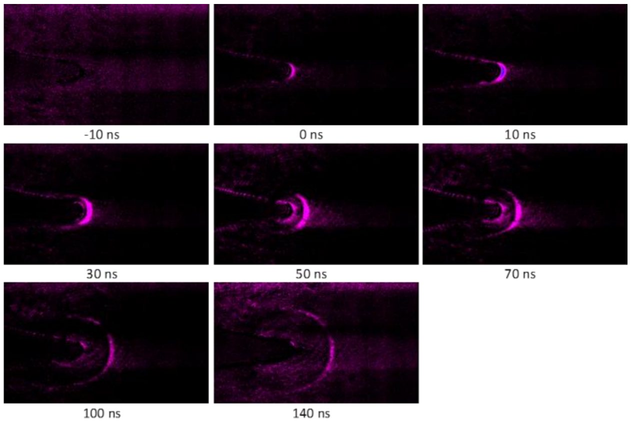

Figure 3: Schlieren imaging of electrostriction wave propagation through pure water. Image size is 750x500um and compression wave velocity is 1.4 0.2 km/s.

When the liquid was changed to the water-ethanol mixture, the same wave is seen to form, even with the reduced dielectric permittivity of the new solution, as seen the pictures of Figure 4. The variation in the contrast between images is a result of differences in camera focuses at the time the experiment was performed. Quantitatively one cannot accurately compare the two cases, but for our purposes, qualitative assessment was the only outcome desired. It was sufficient to observe that the electrostriction wave still propagated even though the estimated value of was lower. In the case when the liquid was pure ethanol, no similar compression wave propagation was seen.

The equation for total pressure in the liquid (1) was solved numerically along with the equations for compressible hydrodynamics of the dielectric fluids under the stress of fast rising pulsed electric fields [7, 8, 10]. The simulations results are shown in Figure 5 for liquid water. In the results, we also the formation of a strongly compressed region immediately with the voltage pulse at the electrode tip, followed by the propagating compression wave away from the electrode.

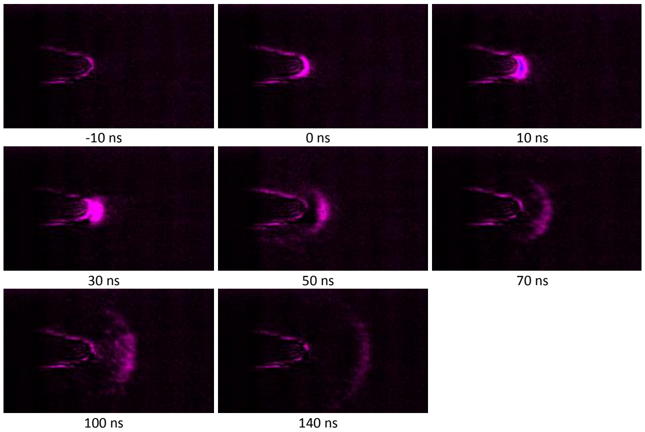

Figure 4: Schlieren imaging of electrostriction wave propagation through the liquid for water-ethanol mixture. Image size is 750x500um and compression wave velocity is 1.5 0.2 km/s.

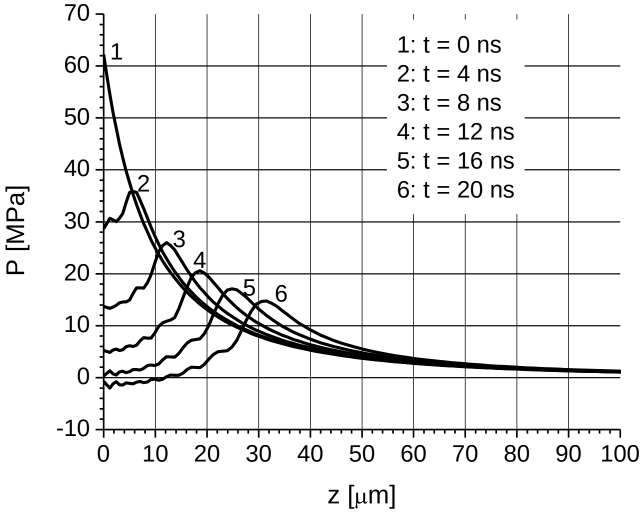

Figure 5: Hydrostatic pressure distribution along the axis of the high voltage electrode in liquid water. 0 m represents immediately near to the electrode tip. The wave propagation speed is ~ 1.5km/s.

4. Discussion

The results from the experiments agree with the simulation results from the electrostriction model. According to the model, if in the water is less than , this leads to density perturbations due to the cavitation effect caused by the negative pressure. As the voltage pulse reaches the plateau, eventually the hydrodynamic forces compensate to balance the negative pressure and leads to a compressed region near the electrodes. Naturally the system will relax and this compressed liquid layer will propagate into the liquid away from the zone of high pressure until the stored energy is dissipated.

In the context of these experiments, the interesting result is not the shockwave which follows, but the processes leading to the formation of this density perturbation. We find some evidence in the possibility of this being a key process to the initiation processes leading to the generation of nanosecond pulsed discharges in liquids. If these electrostriction waves are formed as a result of the formation of "nanopores" near the electrode, then these voids in the liquid could provide the necessary space for electrons to accelerate and cause ionization. The effect is also sensitive to the square of the electric field according to equation (1). If is high enough, this will produce a region much more saturated with these nanopores, as well as deliver much more energy to free electrons to cause ionization – that is, the minimum criteria for avalanche to streamer formation has been met ([6].

5. Conclusion

We have shown that nanosecond-pulsed electric fields applied to submerged electrodes lead to density perturbation in the liquid. These density changes behave in a manner as predicted by the electrostriction model predicted in [7]. Modelling results agree with experimentally obtained Schlieren imaging of the density changes occurring near the electrode tip. The resulting electrostriction wave which propagates through the liquid can be as a result of a region of negative pressure which forms as a consequence of localized nanopores creation near the tip. In this case, we have identified a mechanism via which discharge can be initiated in the liquid phase which does not require heating or bubble expansion.

6. References

1. Graham, W.G. and K.R. Stalder, *Plasmas in liquids and some of their applications in nanoscience.* Journal of Physics D: Applied Physics, 2011. **44**(17): p. 174037.
2. Kolb, J.F., et al., *Streamers in water and other dielectric liquids.* Journal of Physics D: Applied Physics, 2008. **41**(23): p. 234007.
3. Bruggeman, P. and C. Leys, *Non-thermal plasmas in and in contact with liquids.* Journal of Physics D: Applied Physics, 2009. **42**(5): p. 053001.
4. Starikovskiy, A., et al., *Non-equilibrium plasma in liquid water: dynamics of generation and quenching.* Plasma Sources Science and Technology, 2011. **20**(2): p. 024003.
5. Dobrynin, D., et al., *Non-equilibrium nanosecond-pulsed plasma generation in the liquid phase (water, PDMS) without bubbles: fast imaging, spectroscopy and leader-type model.* Journal of Physics D: Applied Physics, 2013. **46**(10): p. 105201.
6. Raizer, Y.P., J.E. Allen, and V.I. Kisin, *Gas Discharge Physics*. 2012: Springer London, Limited.
7. M. N. Schneider, M.P., A. Fridman, *Theoretical Study of the Initial Stage of Sub-nanosecond Pulsed Breakdown in Liquid Dielectrics.* IEEE Transactions on Dielectrics and Electrical Insulation, 2012. **19**(5): p. 1597-1582.
8. Shneider, M.N. and M. Pekker, *Dielectric fluid in inhomogeneous pulsed electric field.* Physical Review E, 2013. **87**(4).
9. Starikovskiy, A., *Pulsed nanosecond discharge development in liquids with various dielectric permittivity constants.* Plasma Sources Science and Technology, 2013. **22**(1): p. 012001.
10. Seepersad, Y., et al., *On the electrostrictive mechanism of nanosecond-pulsed breakdown in liquid phase.* Journal of Physics D: Applied Physics, 2013. **46**(16): p. 162001.