Gas-water two-phase discharge phenomenon in PEF chamber for water treatment

Zhiyuan Wang, Ruobing Zhang and Zhihao Chen
Tsinghua Shenzhen International Graduate School, 518055, Shenzhen, China

Email: zy-wang17@mails.tsinghua.edu.cn

Abstract. Pulsed Electric Field treatment is a critical technology in waste water treatment and sterilization. However, the discharge of the treatment chamber that causes the damage of pulse generators is one of the main technical issues of its applications. Bubbles under water are the most vulnerable sections and could cause breakdown since it is too difficult to generate discharge in the bulk of water under microsecond-pulse and relatively low electric field(<70kV/cm). Uniform discharging images of gas bubbles under water are captured. The influence of γ approach on breakdown in short gas gap (under 1cm) with water electrode under atmospheric pressure is confirmed. The influence of conductivity of aqueous solution and the repetitive frequency of voltage pulses is discussed. It can be confirmed that the breakdown phenomenon of the PEF treatment chamber is mainly caused by air bubbles, which are generated by Joule heat process as well as cavitation and then discharge under the metal cathode. This work presents vital references for improving the design of PEF treatment chambers.

1. Introduction
Pulsed Electric Field (PEF) is widely studied as an innovative waste water treatment and sterilization technology with less energy loss and high efficiency. The electric field is efficient enough to be used as sterilization purpose only if it is higher than 15kV/cm [1].

Electric discharges with liquid have been widely studied both theoretically and for application purpose in the past. Most of researches done by now are mainly focused on two types of discharging. First is the impulse breakdown of the bulk of water, which requires the intensified electric field above 1MV/cm with pin electrode to initiate plasma channel [2-3]. Then although the breakdown of liquid with the aid of bubbles is discussed, the electric field is much higher than PEF condition which is up to 300kV/cm [4]. What’s more, most studies are based on high output power energy (over 100 J/pulse) and single pulses [5-6]. In the meanwhile, the PEF pulses are short pulses with higher repetitive frequency and lower energy.

Compared with gas, the liquid has a higher density and the mean free path of electron is shorter, so the breakdown field strength of the liquid is usually above 10 MV/cm [7]. In order to achieve this particular of electric field strength, the current research on liquid breakdown mechanism is mostly carried out under the condition of extremely uneven electric field such as the needle to plate electrode. In liquid containing dissolved gases or bubbles, it is generally believed that liquid breakdown starts from bubble breakdown [8]. The PEF treatment requires a uniform electric field and the field strength is usually below 70 kV/cm [9]. The complete breakdown of the PEF treatment chamber found in this paper is related to the bubble breakdown in the water.
The breakdown phenomena within bubbles under water appeals wide attentions from scholars. The impact of air bubbles is studied with numeric simulations which suggest the minimum breakdown strength for air bubbles is about 30kV/cm [10], but it is not confirmed whether the isolated bubbles under water are able to discharge in experiment. Although the plasma inside water bubbles had been acquired with a needle electrode [11], the gas is highly likely pre-ionized before it comes into water. Discharging within isolated bubbles is also found in literature [12], but the experiments was also performed with a pin to plate configuration as the bubble is extremely close to electrode tip.

In general, the PEF treatment chamber are usually designed to form uniform electric field to ensure the treatment result. Most discharging phenomenon of bubbles found by now are performed with nonuniform electric field strength much stronger than normal PEF treatment level. Breakdown of PEF treatment chamber more resembles to discharge within gas bubbles under repetitive pulses. Pulse parameters are found related to the breakdown strength of air under atmospheric pressure [13]. Considering in the practical PEF treatment, the treated liquid is conductive aqueous solution, so the influence of conductivity of water as electrode on breakdown strength is also studied in this paper.

2. Experiment results and discussion

2.1. Experiment setup

This experiment uses a high voltage pulse power generator with adjustable parameters. The pulse generator generates pulse voltage with amplitude continuously adjustable from 0-20 kV, and can be adjusted to unipolar or bipolar output with a pulse rise time of 50 ns. The experiment is performed inside a cylinder bottle (inner diameter 90mm) with two plate electrodes made of SUS316 (diameter of the flat bottom 40mm). The gap distance between the electrodes is adjustable.

The applied voltage and loop current are captured respectively by a high-voltage probe (Tektronix P6015A) and current monitor (Pearson Model 411) coupled to a digital oscilloscope (Agilent DSO7104A).

The breakdown strength of gas gap is tested with aqueous electrode under unipolar repetitive pulses. The experiment configuration is as Figure 1 shows. Injecting 150ml water of different conductivities to the container, changing the gas gap distance between upper electrode and water surface as well as the parameters of repetitive pulses generated, it is able to test the breakdown strength of air between metallic cathode and water anode at the atmospheric pressure.

![Figure 1. Schematic diagram of experiment setup.](image)

2.2. Gas gap breakdown with water phase electrode

Figure 2 shows the breakdown voltage of the gas gap (d=2.8mm) changes as the repetitive frequency and the conductivity of water varies. It is found that under relative low conductivity (<200μS/cm) the breakdown voltage doesn’t show great differences. When conductivity and frequency increase, the breakdown voltage seems to drop because of the greater electric force causes vibrations on the surface of water.
**Figure 2.** The influence of water conductivity and pulse frequency on breakdown voltage.

Figure 3 shows the breakdown voltage changes as the gas gap distance changes when the water conductivity is 200μS/cm. When the gas distance is 1.8mm, 2.8mm and 3.8mm, the breakdown strength is about 44.4 kV/cm, 39.3kV/cm and 35.5 kV/cm. The average breakdown strength is dropping as the gas gap becomes longer. The typical discharging images are as Figure 4 shows. It is widely known that the minimum breakdown strength of air is about 30kV/cm under atmospheric pressure, but in experiments it can be seen under 5mm the average breakdown strength is above 30kV/cm.

**Figure 4.** Discharging images between water anode and stainless-steel cathode under repetitive voltage pulses.

Since the PEF processing is typically at atmospheric pressure, and the spacing of PEF treatment chamber’s electrodes is typically less than 1 cm (pd < 100 kPa.cm), which is not sufficient long to form the critical electron avalanche. Therefore, the air gap breakdown within 1 cm under atmospheric pressure should be explained by Townsend’s discharge theory, which emphasizes the importance of the function of cathode and it is consistent with the experiments that show the difficulty to ionize a totally isolated floating bubble. The water surface decreases the contribution of secondary electrons to swarm if it serves as cathode. Even with a pin to plate experiment setup an isolated bubble has been successfully initiated, but the electric field at the pin electrode is 1.2MV/cm [14], which is not the situation that happens under PEF treatment condition. Hence this paper focus on the scenario that bubbles are attached to metallic electrode. If the cathode is metal electrode, which has a higher secondary electron emission factor $\gamma$, the breakdown phenomena is most reproducible and the breakdown voltage is always the lower than when it is aqueous solution. The emission of electrons from aqueous solution is related to the electrochemical reactions at the plasma-liquid interfaces discussed in the literature [15].
2.3. Gas bubble breakdown under water
Since most bubbles shape as ellipsoids and it is found that under uniform external electric field, any ellipsoid will maintain its internal electric field uniform, and this property is irrelevant with the shape of the ellipsoid in the literature [16]. However, it is too difficult to acquire desired bubbles with specific parameters, an equivalent experiment is conducted to confirm that the bubbles attached to the plate are subjected to uniform electric field.

Helium bubbles are injected into the water between the parallel plate electrodes in the experiment at Figure 5 and 6. It is found that uniform discharge could appear in helium of which has lower breakdown strength at atmospheric pressure. In order to acquire uniform discharge it is needed to decrease the \( \alpha/p \) (\( \alpha \) is the electron impact ionization factor which is related to electric field strength) [17], and the only way to make it at atmospheric pressure is to reduce the breakdown strength.

High speed camera (Phantom V2012) with maximum film rate 100000 fps and LED backlight are used. The gap distance of plate is 4mm. Deionized water (conductivity 1μS/cm) is used. The magnitude of unipolar pulse is about 10kV, pulse width 50μs, repetitive frequency is 50Hz. Since the minimum exposure time of the high-speed camera is 9.4μs at the resolution of 256×256. It can be found out that the bubbles inside water appear to discharge in a transition way from glow to streamer discharge, suggesting that the electric field inside underwater bubble is uniform.

Figure 6 shows typical consecutive frames captured at 10μs intervals just before and after electrical discharge occurred. The image at 0μs represents a control frame without electrical discharge in bubble. While in the next frame glow like discharge is captured, and it transfers from glow to streamer in the following two frames at 20μs and 30μs. Glow in transition to streamer discharges are also found at subsequent pulses with an interval of 19.5ms, although the bubble surface with vibrations is not as regular as ellipsoids.

2.4. Discussion of experiment results
The breakdown phenomenon of gas gap above water and gas bubbles inside water are mathematically equivalent. First, the electric field of gas phase in the experiment is all uniform. It could be seen from Figure 6 that uniform discharge also appears at consecutive voltage pulses, and the uniformity of the electric field is not severely affected by the shape of gas bubble. Therefore, the breakdown voltage in gas bubble could also be explained by the Paschen’s law. Secondly, the length of the bubble and the
gas gap are usually less than 1 cm, so the Townsend’s discharge theory could explain the development of discharge and the breakdown strength. What’s more, as Figure 5 and 6 shows, uniform discharging is captured, which is the typical phenomenon of Townsend’s $\gamma$ approach. Finally, due to the great difference of the dielectric constant of water and gas, the breakdown is initiated in gas phase and develops between the electrodes of the treatment chamber.

3. Conclusions
In summary, it is found that under PEF treatment condition, the breakdown strength of short air gap with water phase electrode as anode is rather stable with aqueous solution of low conductivity (<50 $\mu$S/cm) under low pulse repetitive frequency (f<1000Hz). The breakdown strength is above 30kV/cm, which is above the normal breakdown strength considered in most relative researches, and increases as the gas gap decreases when the gas gap length is shorter than 5 mm.

In the experiment, under-water bubbles are broken with plate electrode and it is first seen in this paper that bubbles are discharging uniformly. This experiment has two significant meanings. First it is concluded that under uniformed external electric field the bubbles under water are also subjected to uniformed electric field. With the great different of the dielectric constant between water and gas, the voltage applied is mostly fallen on the gas section, which causes the gas phase first to discharge. Then the gas phase discharging initiates the breakdown of the whole treatment chamber. What’s more, the uniform discharging found in this experiment, which is a typical phenomenon of the influence of $\gamma$ approach, confirms that the breakdown mechanism of bubbles under water should be explain by the Townsend’s theory of discharge.

In conclusion, it is explained why breakdown happens at a relatively low electric field strength in PEF treatment chambers. In addition, the influence of pulse parameters and conductivity of water on breakdown strength of gas bubbles is studied. It is given a clear idea that the discharge under PEF treatment condition is a gas discharging initiating breakdown phenomenon should be explained by Townsend’s discharge theory. This paper provides vital references in designing PEF chambers for waste water treatment and for sterilization propose.

References
[1] Uchida S, Houjo M, Tochikubo F 2008 Journal of Electrostatics 66 427
[2] Clements J S, Sato M, Davis R H, 1987 IEEE Transactions on Industry Applications 2 224
[3] Jones H M, Kunhardt E E 1994 IEEE Transactions on dielectrics and electrical insulation 11 1016
[4] Korobeinikov S M, Melekhov A V, Besov A S 2002 High temperature 40 652
[5] Schneider M N, Pekker M 2015 Journal of Applied Physics 117 224902
[6] Schneider M N, Pekker M 2013 Physical Review E 87 043004
[7] Seepersad Y, Pekker M, Schneider M N, et al. 2013 Journal of Physics D: Applied Physics 46 162001
[8] Bruggeman P, Leys C, Vierendeels J 2007 Journal of Physics D Applied Physics 40 1937
[9] Zhang R, Zheng N, Liu H, et al. 2015 IEEE Transactions on Plasma Science 43 610
[10] Góngora-Nieto M.M, Pedrow P D, Swanson B G, Barbosa Cánovas G V 2003 Innovative Food Science & Emerging Technologies 4 57
[11] Tachibana K, Takekata Y, Mizumoto Y, Motomura H, Jinno M 2011 Plasma Sources Science and Technology 20 034005
[12] Bruggeman P J, et al. 2016 Plasma sources science and technology 25(5) 053002
[13] Shao T, et al. 2005 Proc. Chin. Soc. Electr. Eng. 25 161
[14] Foster J E, Sommers B, Gucker S 2014 Japanese Journal of Applied Physics 54 01AF05
[15] Akolkar R, Sankaran R M 2013 Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films 31 050811
[16] Landau L D, Bell J S, Kearsley M J, et al. 2013 Electrodynamics of continuous media 39
[17] WANG X X 2009 High Voltage Eng. 35 1