Electrical Breakdown in Liquids

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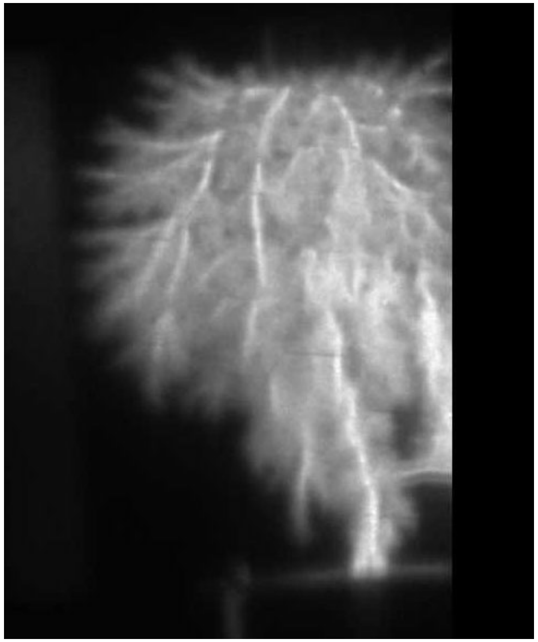
Kyoung-Jae Chung

Department of Nuclear Engineering

Seoul National University

Water spark gap switch

- The use of water insulated switches to transfer energy from one stage to another in large pulsed power drivers has led to a high degree of integration not possible with gas switches.
- This, coupled with the relatively high breakdown strength of fast charged water systems, allows for short gap lengths and correspondingly lower switch inductance.



Discharges in and in contact with liquids

- Discharges in and in contact with liquids provide new scientific challenges and emerging technological opportunities for the plasma community. e.g) plasma surgery, lithotripsy, plasma oncology, decontamination and purification of water.
- Discharges in and in contact with liquids generate intense UV radiation, shock waves and active radicals (OH, atomic oxygen, hydrogen peroxide, etc) all of which are effective agents against many biological and chemical matter.

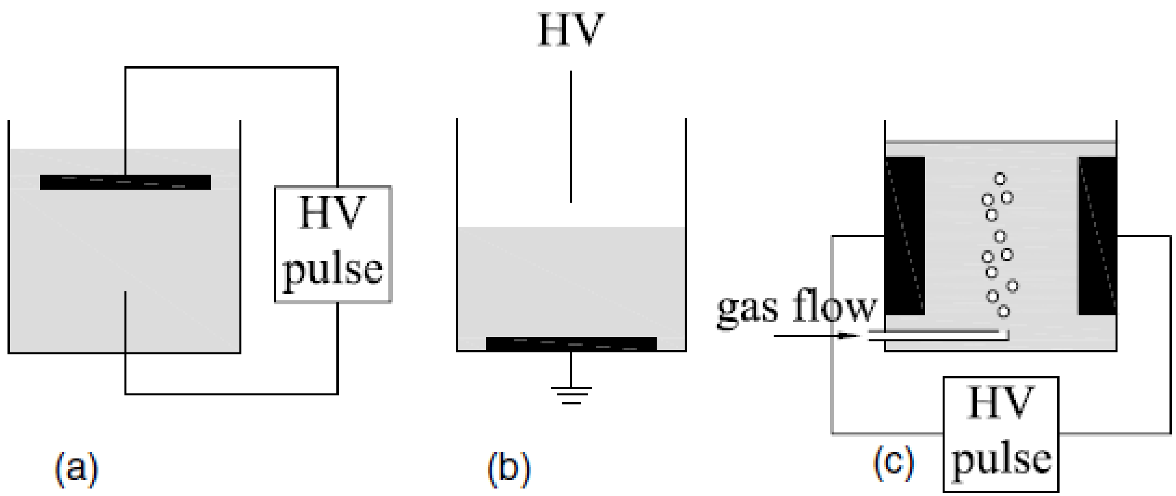


Figure 1. Typical electrode configurations for the three different types of discharges in and in contact with liquids. (*a*) direct liquid phase discharge reactor (*b*) gas phase discharge reactor with liquid electrode (*c*) example of bubble discharge reactor.

Liquid breakdown has no single comprehensive theory

- The breakdown mechanism in liquids is more complicated than in solids or gases since liquids are much denser in comparison with gases, and do not exhibit the long range order seen in solids.
- Additionally the purity of the liquid, such as dissolved gases which form micro-bubbles in the liquid, plays a significant role in the breakdown process.

- Two categories for liquid breakdown:
  - Electronic mechanism (direct impact ionization): The discharge is a consequence of avalanche multiplication of free charge carriers in the liquid and is described by the gas discharge model generalized to the liquid phase. It implies that electrons in strong fields are accelerated in liquids and ionize molecules and atoms.
  - Bubble mechanism: Discharge occurs in gas cavities either already presented in the liquid and on the electrodes or newly formed under voltage exposure.
- A general acceptance is growing that pre-existing bubbles and field enhancement effects in the near electrode region are involved even for nanoseconds voltage pulse widths.

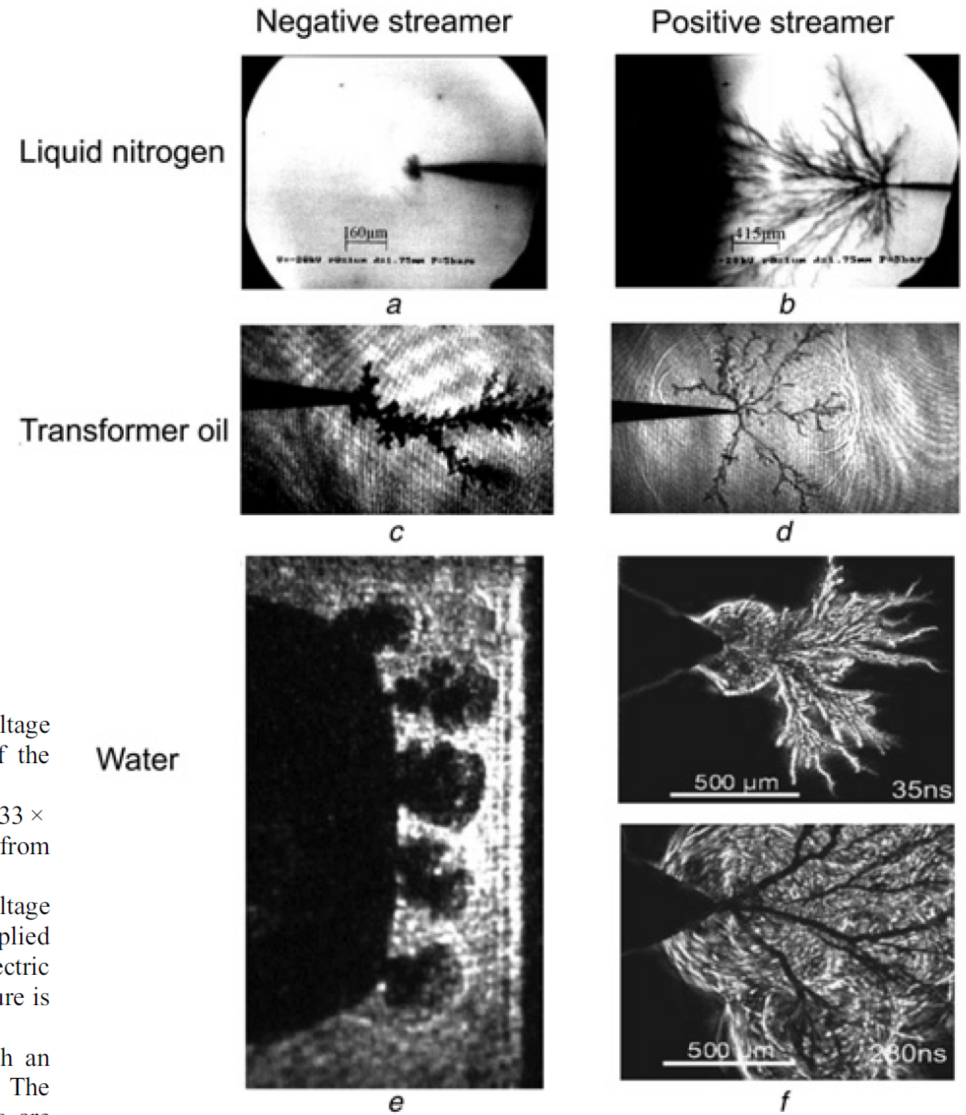
R. P. Joshi, Plasma Chem Plasma Process 33, 1 (2013)

Universally observed characteristic features during liquid breakdown

- The breakdown strength of fluids increases with increasing pressure.
- The time lag to breakdown increases with increasing pressure.
- The breakdown voltage is always lower when the polarity of the stressed electrode is positive than when it is negative. This means that when a symmetrical electrode system is used pre-breakdown is preferentially initiated at the anode.
- The time lag to breakdown is polarity dependent. A formative time lag for micro-bubble formation is observed for negative streamer initiation and not for positive streamers.
- The critical electrical breakdown field decreases with increasing pulse width.
- The probability of breakdown increases with increasing surface area of the electrode. This is correlated with the increased probability of the existence of imperfections like micro-protrusion on the electrode surface which are able to locally enhance the electrical field or cause field emission to trigger breakdown.

Polarity effect: positive streamer vs. negative streamer

- Positive streamer:
  - Thin and filamentary structure
  - Faster initiation than negative mode
  - Initiate at lower voltage
- Negative streamer:
  - Thick root, bush-like or mushroom-like structure
  - Slower initiation than positive mode
  - Initiate at higher voltage



a, *b* Present streamers formed in a 1.75 mm gap under different applied voltage (−20 and +28 kV) in liquid nitrogen. The pressure is 5 bar and the radius of the needle electrode is 1 µm. The figure is reproduced from [46]
c, *d* Present streamers formed in a 2.5 mm gap in transformer oil. The pressure is 1.33×10^{-5} bar and the radius of the needle electrode is 20 µm. The figure is reproduced from [47]
e Shows negative streamers from a needle cathode in water when a rectangular voltage pulse of 200 ns (full width half maximum) with a slow rise time of about 80 ns is applied in a 400 µm gap. The radius of the needle electrode is 0.8 mm and the maximum electric field close to the needle electron is of 1.4 MV/cm. The pressure is 1 bar. The figure is reproduced from [34]
f Shows positive streamers in water from a needle anode. A voltage pulse with an amplitude of 18 kV and a step rising time of 40 ns is applied in a 20 mm gap. The radius of the needle electrode is 25 µm and the pressure is 1 bar. The figures are reproduced from [48]

A. Sun, High Volt. 1, 74 (2016)

Determination of breakdown delay time (time lag)

- For a large number of experiments:

$$N(t) = N_0 \exp\left[-\frac{t - t_f}{t_s}\right]$$

$$\tau_{bd} = t_s + t_f$$

  - $N(t)$: the number of pulses for which the breakdown time lag is equal or larger than t.
  - N_0: the total number of breakdowns.
  - t_s: statistical delay time.
  - t_f: formative delay time.
  - τ_{bd}: total breakdown delay time (breakdown time lag).

- Laue plot

$$-\ln N(t)/N_0 = \frac{t - t_f}{t_s}$$

V. Ushakov, Impulse Breakdown of Liquids (2007)

Statistical analysis of breakdown time lag in water

Average = 55 ns
SD = 18 ns

t_s = 16 ns
t_f = 42 ns

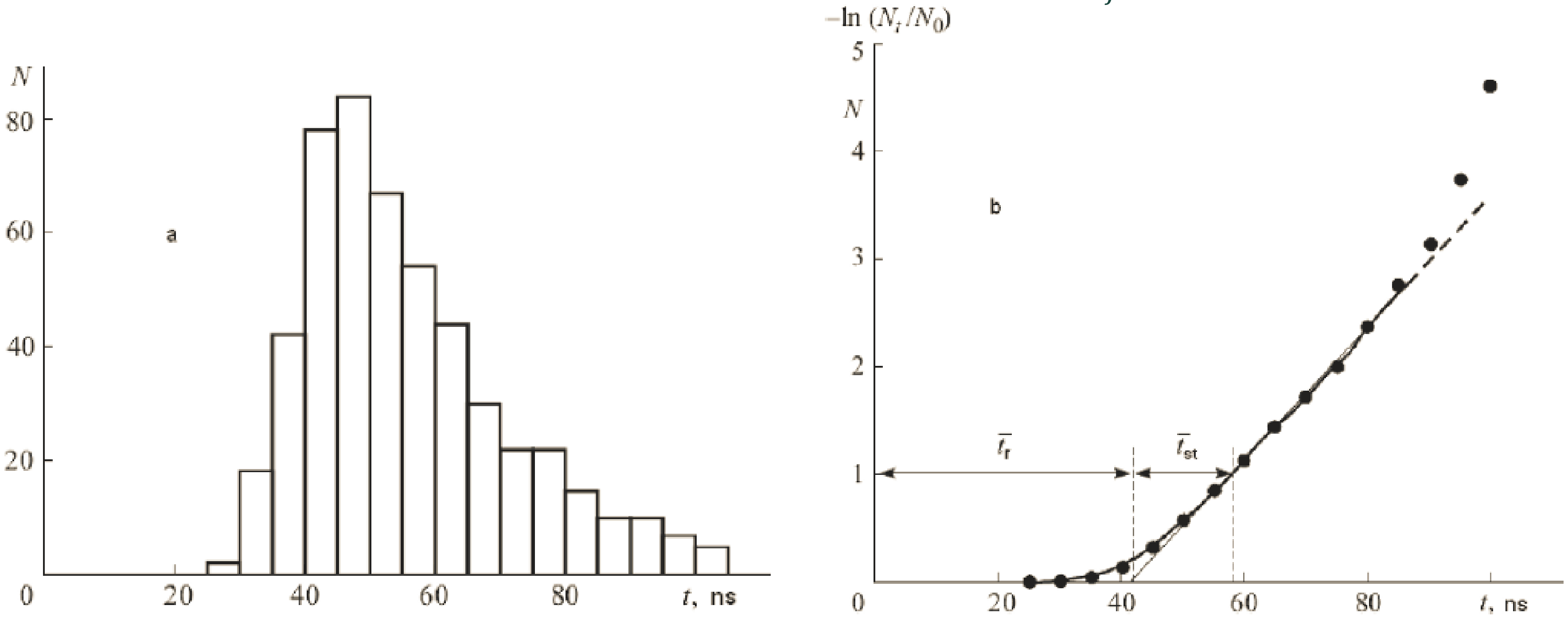


Fig. 5.4. The statistical analysis of breakdown time lag in distilled water: **a)** histogram of the measurements breakdown time lag ($N_0 = 510$) and **b)** breakdown time lag distribution in the Laue coordinates. The electric field strength was 0.6 MV/cm, and the interelectrode distance was 0.2 cm

The time lag to breakdown (τ_{bd}) increases with increasing pressure

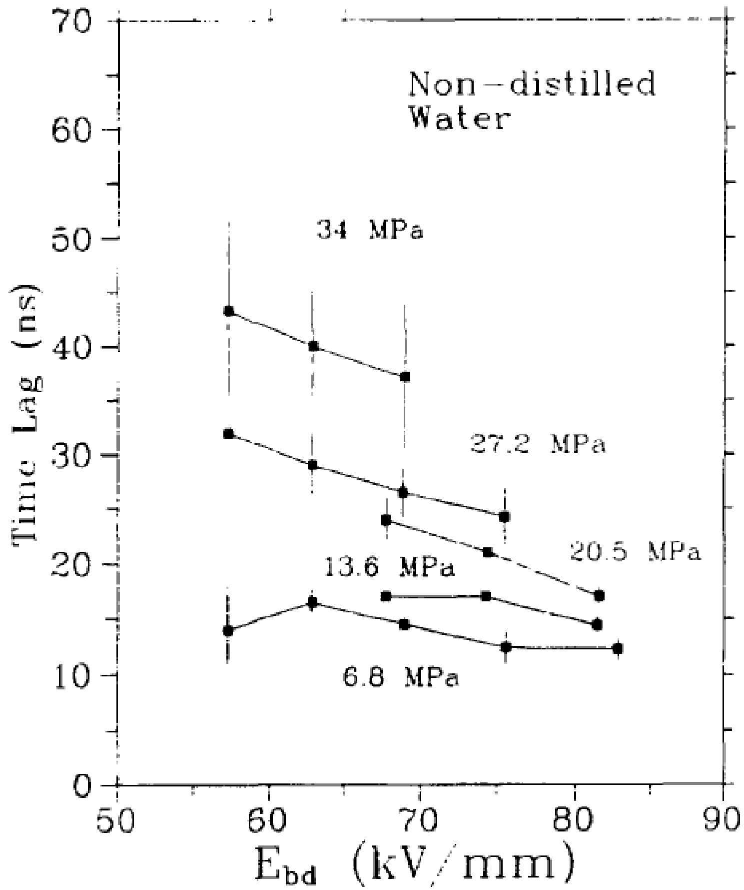


Figure 5.

Dependence of τ_{bd} in water (d = 1.1 mm) as a function of applied field with pressure as parameter for de-ionized, non-distilled water. Note the increase of τ_{bd} with pressure.

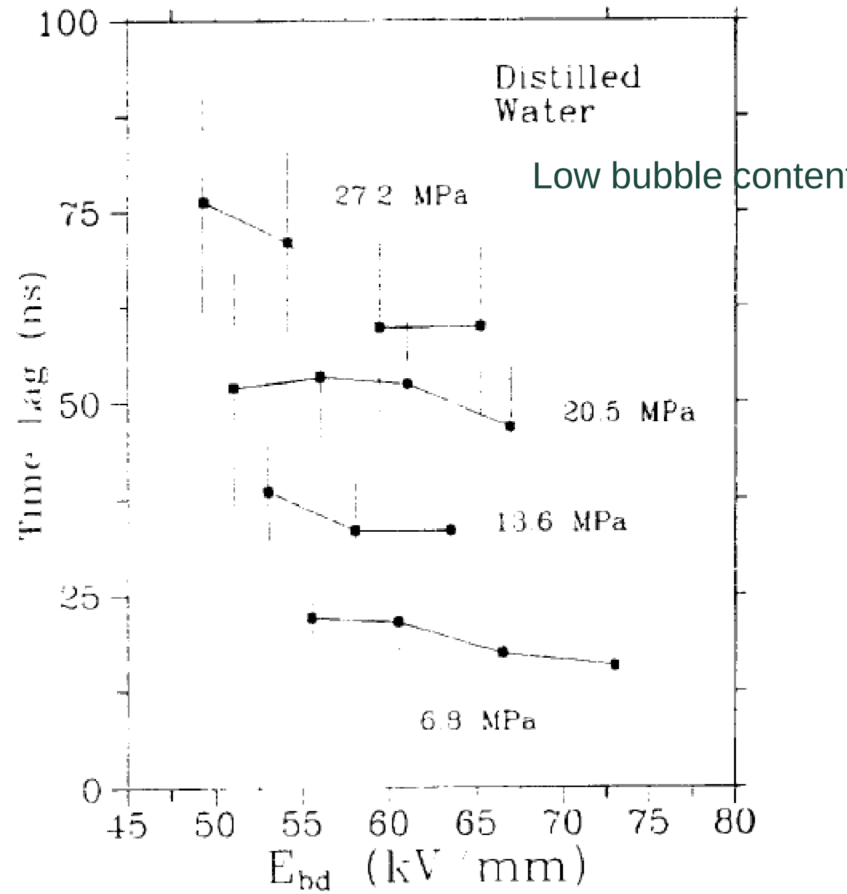


Figure 6.

Dependence of τ_{bd} in water (d = 1.1 mm) as a function of applied field with pressure as parameter for de-ionized, distilled water. Note the increase of τ_{bd} with pressure.

H. Jones, IEEE Trans. Dielectrics and Electrical Insulation 1, 1016 (1994)

The time lag to breakdown (τ_{bd}) increases with increasing pressure

- The strong influence of the external pressure on the statistical delay indicates that the thermal mechanism of microbubble formation is dominant.
- The statistical delay of the electric discharge from anode includes the process resulting in fast local superheating of the liquid near the electrode surface and nucleation.

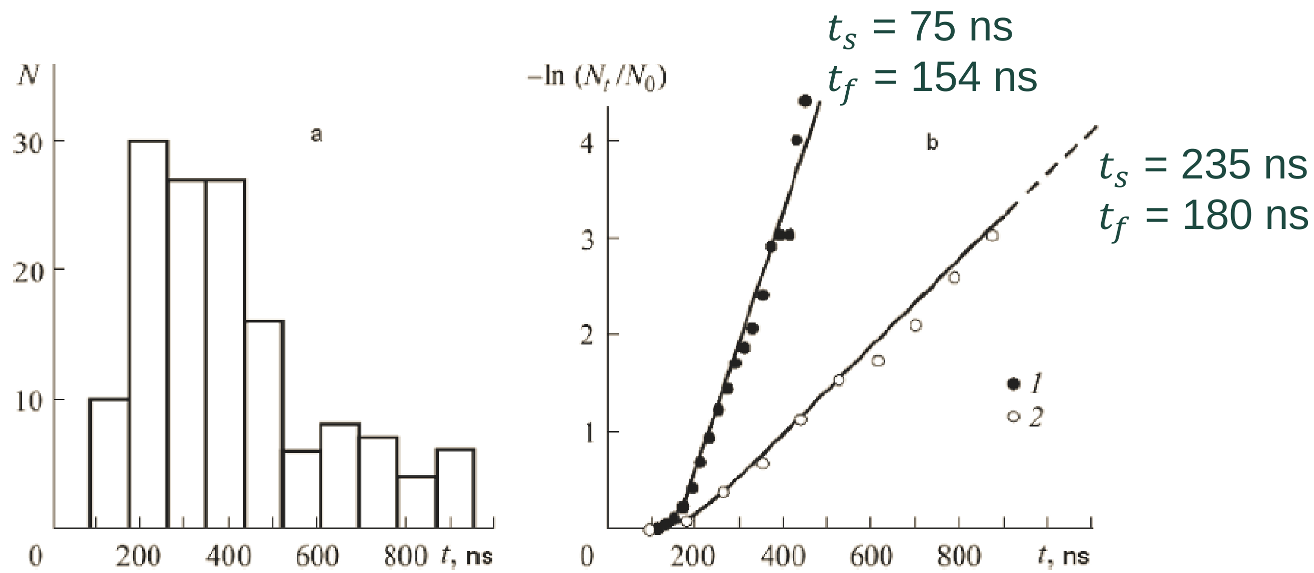


Fig. 5.9. Results of statistical analysis of the breakdown time lag in distilled water: a) histogram of the measured breakdown time lag (the number of measurements was $N_0 = 150$ and $P = 0.75$ MPa) and b) distribution of the breakdown time lag in the Laue coordinates (*curve 1* is for $N_0 = 150$ and $P = 0.1$ MPa and *curve 2* is for $P = 0.75$ MPa). The electric field strength was 0.9 MV/cm, the interelectrode distance was 200 μm

V. Ushakov, Impulse Breakdown of Liquids (2007)

Polarity effect (with or w/o bubble)

Anode initiation

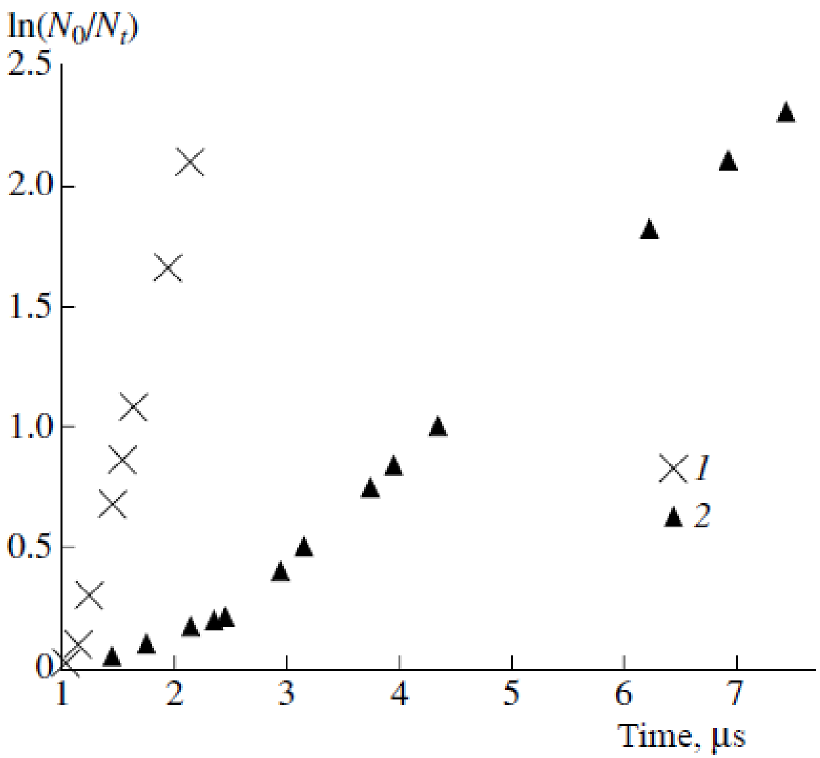


Fig. 1. Breakdown time lag from the anode in Laue coordinates, (*1*) in the presence and (*2*) in the absence of visible bubbles.

Cathode initiation

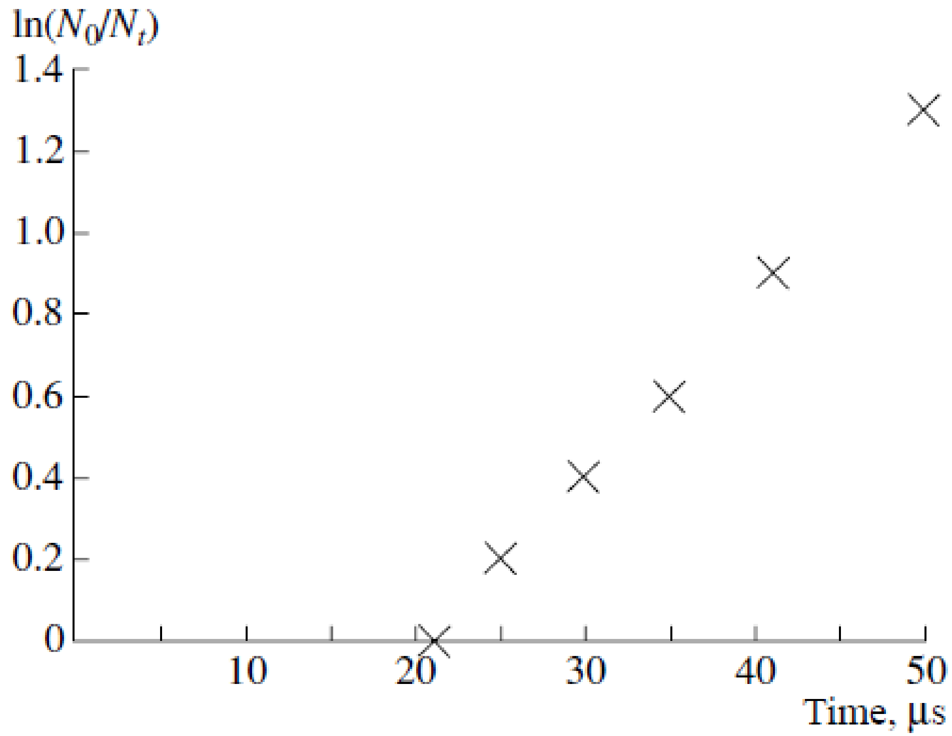


Fig. 2. Breakdown time lag from the cathode in Laue coordinates in the presence of visible bubbles.

S. Korobeinikov, High Temperature 40, 652 (2002)

Polarity effect with bubble

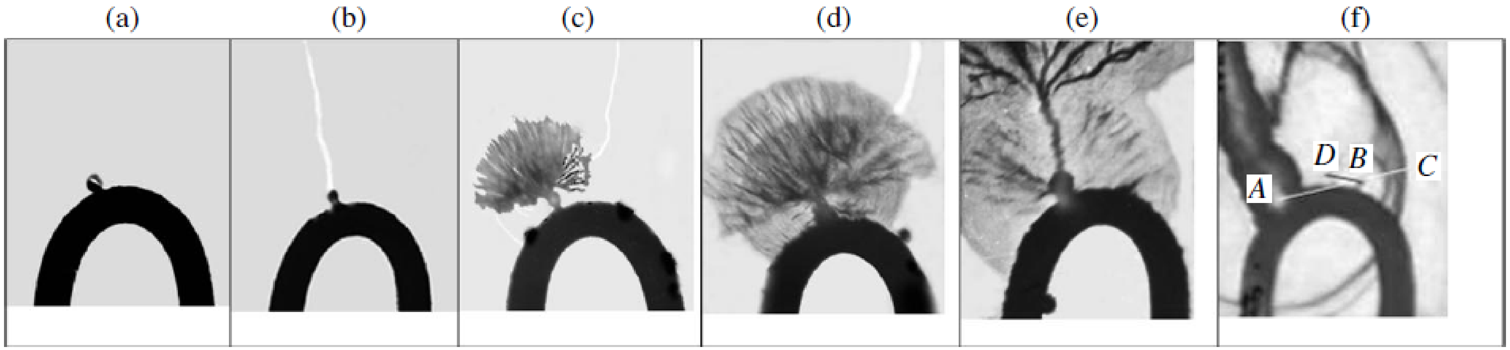


Fig. 3. The sequence of events leading to a breakdown from the anode: (a) initial bubble; (b) deformed bubble; the moment of photographing $\tau_1 = 0.45$ µs after the application of voltage; the moment of discharge $\tau_b = 1.2$ µs after the application of voltage; (c) fan (brush) of streamers from the bubble tip, $\tau_1 = 1.0$ µs, and $\tau_b = 1.3$ µs; (d) fan of streamers before its stopping, $\tau_1 = 1.2$ µs, $\tau_b = 1.4$ µs; (e) disappearance of the first fan of streamers and the emergence of the subsequent fan of streamers, $\tau_1 = \tau_b = 1.5$ µs; (f) postbreakdown hydrodynamics, $\tau_1 = 1.7$ µs, and $\tau_b = 1.2$ µs.

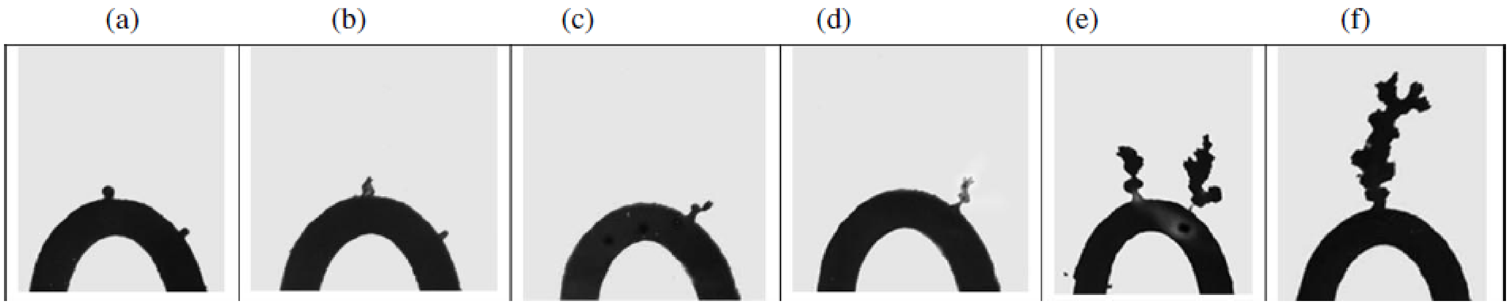
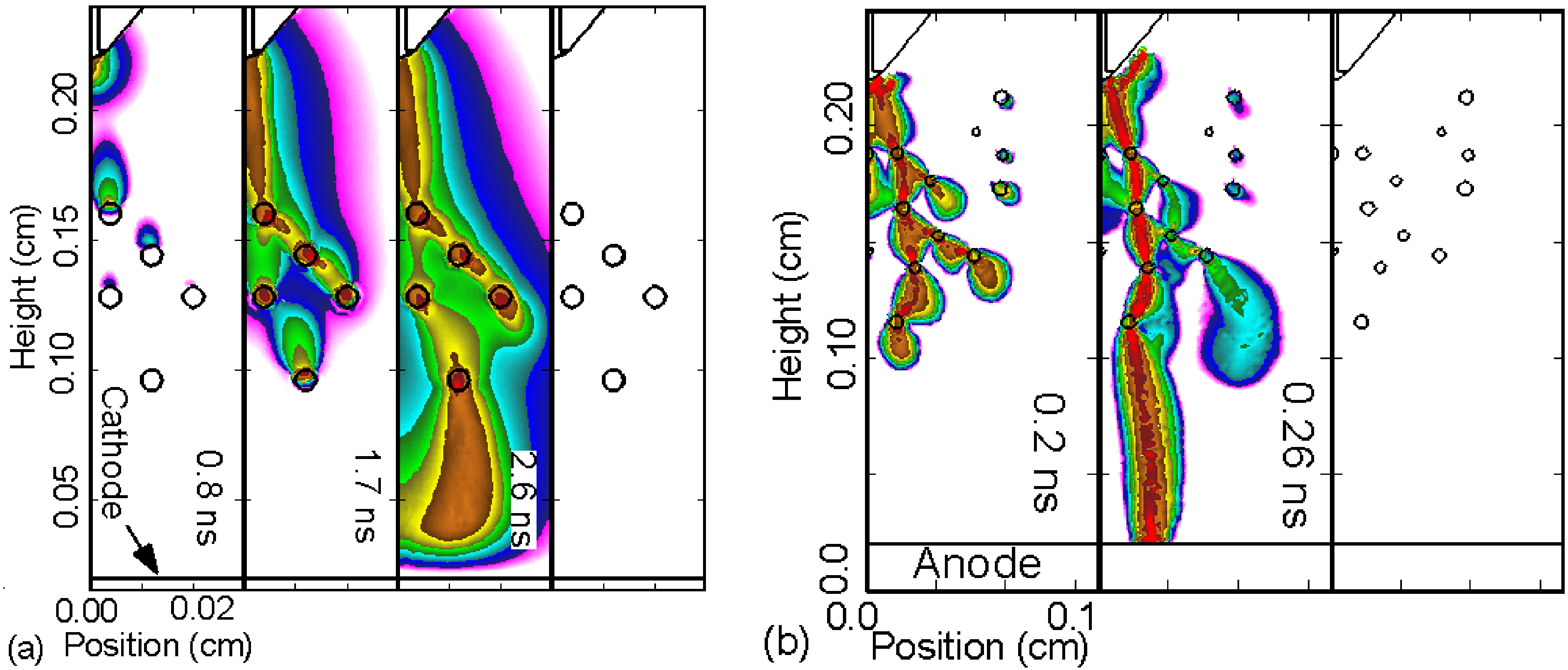


Fig. 4. The sequence of events leading to a breakdown from the cathode: (a) initial bubbles; (b) deformed bubbles, the smaller one is elongated, a bush-like formation begins to grow on the surface of the larger one, $\tau_1 = 0.25$ µs; (c) bubble assumes a "mushroom shape with an elongated thin stem and a cap," $\tau_1 = 0.65$ µs; (d) bubble of finished mushroom shape, with a bush-like formation developing on the bubble surface, $\tau_1 = 0.65$ µs; (e) several bubbles on the surface, a larger "bush" grows from the smaller bubble, $\tau_1 = 1.0$ µs; (f) "bush" of the maximal size, $\tau_1 = 1.5$ µs.

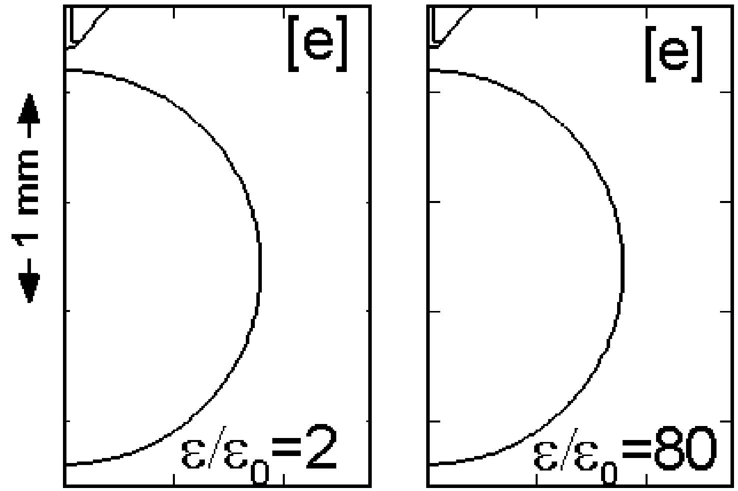
Streamer in water with bubbles

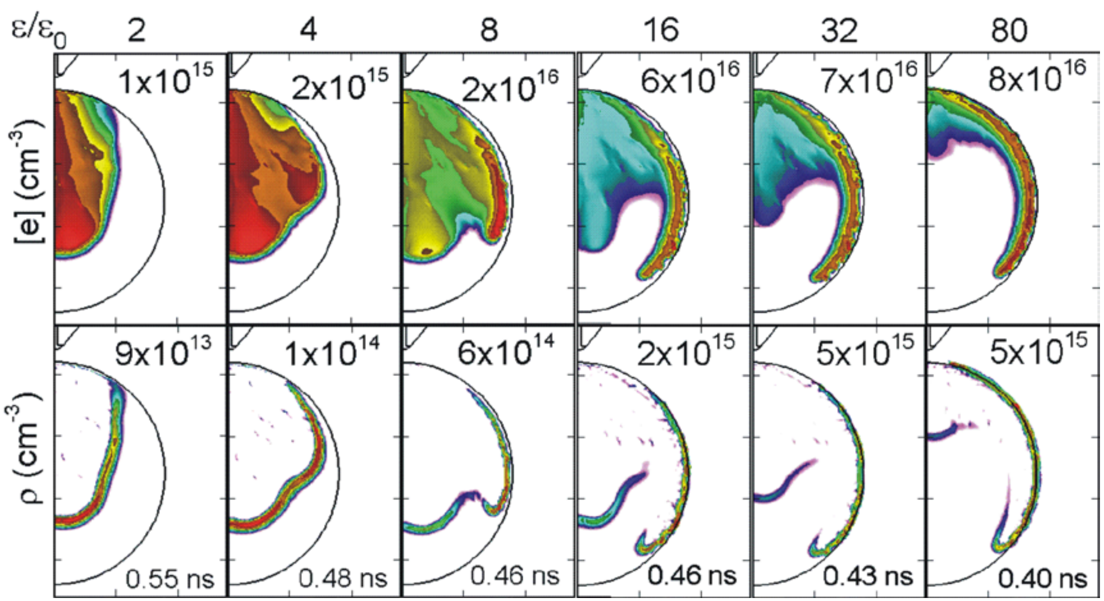


Cathode-directed streamer

Anode-directed streamer

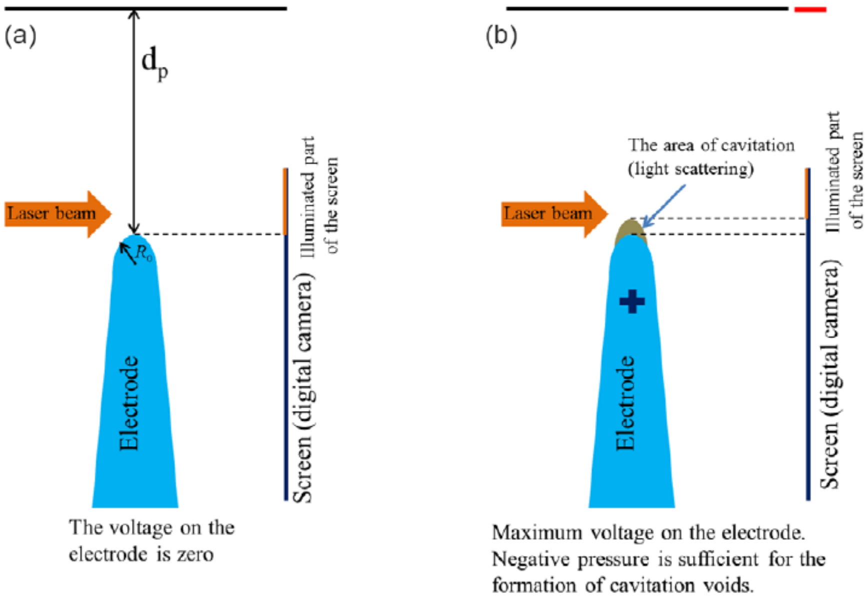
Structure of positive streamers inside bubbles





Formation of cavitation due to electrostriction force

- The initial stage of development of a nanosecond breakdown in liquids is associated with the appearance of discontinuities in the liquid (cavitation) under the influence of electrostriction forces.



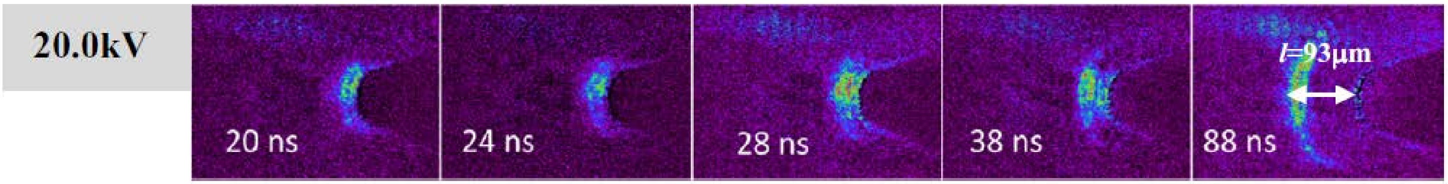
$$\frac{\partial \rho}{\partial t} + \nabla(\rho \vec{u}) = 0$$

$$\rho\left(\frac{\partial \vec{u}}{\partial t} + (\vec{u}\cdot\nabla)\vec{u}\right) = -\nabla p + \frac{\varepsilon_0}{2}\left(\frac{\partial \varepsilon}{\partial \rho}\rho\right)\nabla E^2$$

$$p = (p_0 + B)\left(\frac{\rho}{\rho_0}\right)^{\gamma} - B$$

$$\rho_0 = 1000\,\mathrm{kg\,m^{-3}}, \qquad p_0 = 10^5\,\mathrm{Pa}, \qquad B = 3.07\times10^8\,\mathrm{Pa}, \qquad \gamma = 7.5.$$

$$-\nabla p + \frac{\varepsilon_0}{2}\left(\frac{\partial \varepsilon}{\partial \rho}\rho\right)\nabla E^2 = -\nabla\left(p - \frac{1}{2}\alpha\varepsilon_0\varepsilon E^2\right) = -\nabla p_{\mathrm{total}}.$$



M. Pekker, J. Phys. D: Appl. Phys. 47, 025502 (2014)

Recent research direction

Plasma physics of liquids—A focused review

Patrick Vanraes and Annemie Bogaerts
PLASMANT, Department of Chemistry, University of Antwerp, Universiteitsplein 1, 2610 Wilrijk-Antwerp, Belgium

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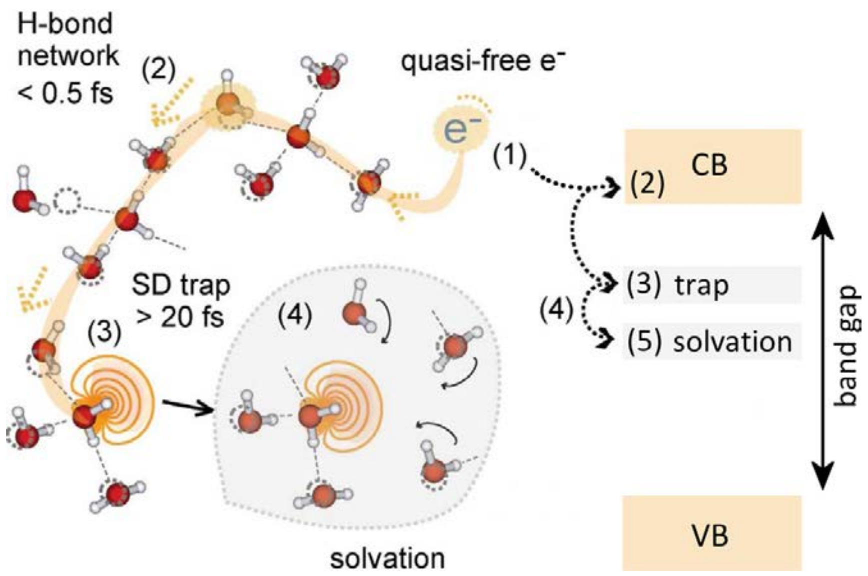
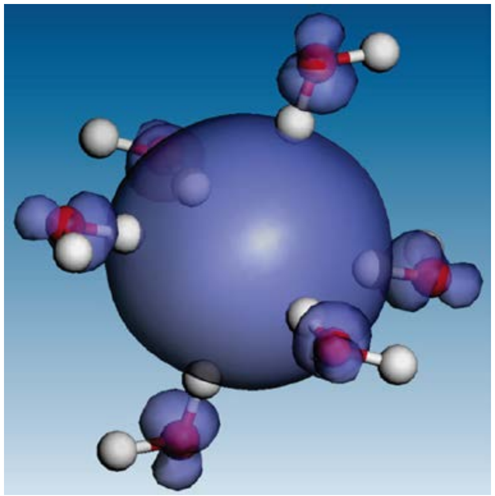
The interaction of plasma with liquids has led to various established industrial implementations as well as promising applications, including high-voltage switching, chemical analysis, nanomaterial synthesis, and plasma medicine. Along with these numerous accomplishments, the physics of plasma in liquid or in contact with a liquid surface has emerged as a bipartite research field, for which we introduce here the term "plasma physics of liquids." Despite the intensive research investments during the recent decennia, this field is plagued by some controversies and gaps in knowledge, which might restrict further progress. The main difficulties in understanding revolve around the basic mechanisms of plasma initiation in the liquid phase and the electrical interactions at a plasma-liquid interface, which require an interdisciplinary approach. This review aims to provide the wide applied physics community with a general overview of the field, as well as the opportunities for interdisciplinary research on topics, such as nanobubbles and the floating water bridge, and involving the research domains of amorphous semiconductors, solid state physics, thermodynamics, material science, analytical chemistry, electrochemistry, and molecular dynamics simulations. In addition, we provoke awareness of experts in the field on yet underappreciated question marks. Accordingly, a strategy for future experimental and simulation work is proposed. *Published by AIP Publishing*. https://doi.org/10.1063/1.5020511

P. Vanraes, Appl. Phys. Rev. 5, 031103 (2018)

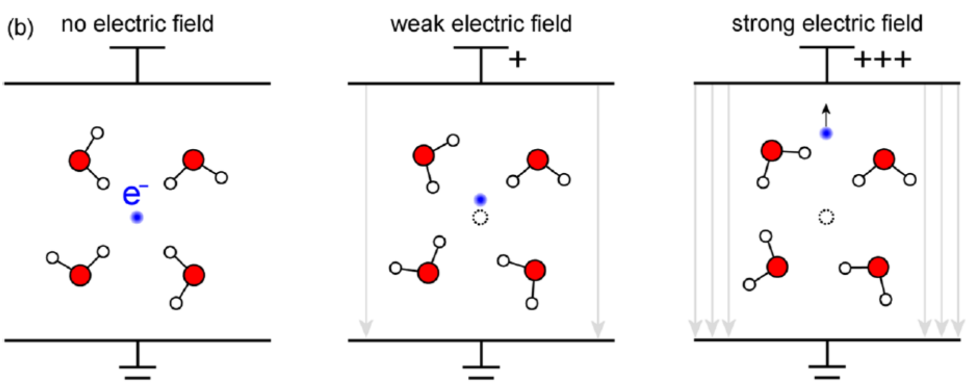
Recent research direction

Electron solvation in water

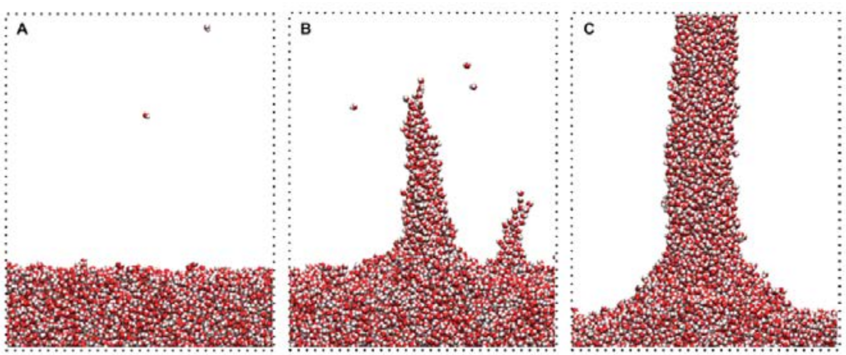




Solvated electron in water



Field-induced desolvation



Surface deformation by electric field

Practical consideration: liquid insulation

- Insulation is the prevention of current flow. It is the major technological problem of high-voltage pulsed power applications.
- There is no simple theory of breakdown in solids and liquids. Knowledge of insulating properties is mainly empirical. These properties vary considerably with the chemical purity and geometry of the insulating material.
- The pulsed voltage dielectric strength [MeV/cm] of transformer oil is approximately

$$E_{\max} \cong 0.5/t_p^{0.33} A^{0.1}$$

The surface area of the high voltage electrode (cm^2)

The time during which the voltage is above 63% of the maximum value (μs)

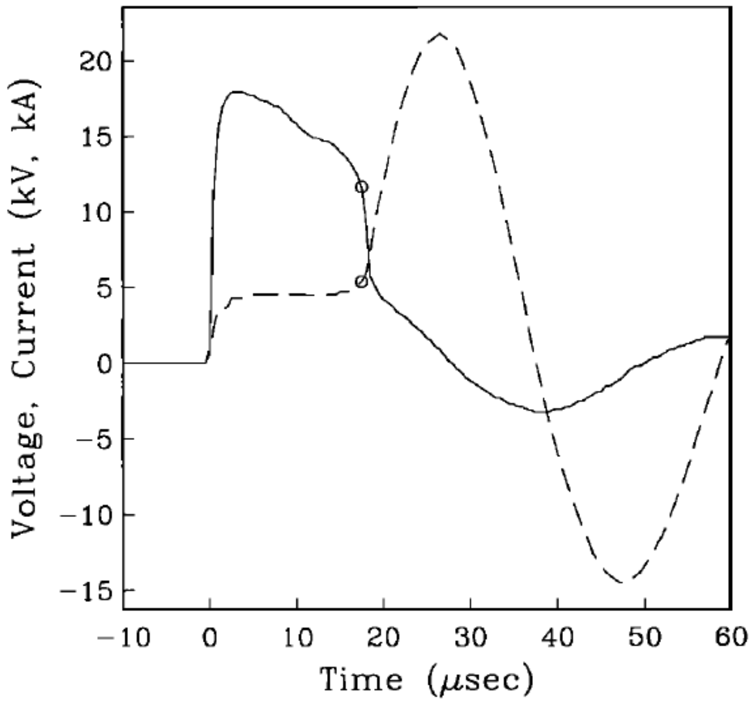
- The dielectric strength of water is dependent on polarity:

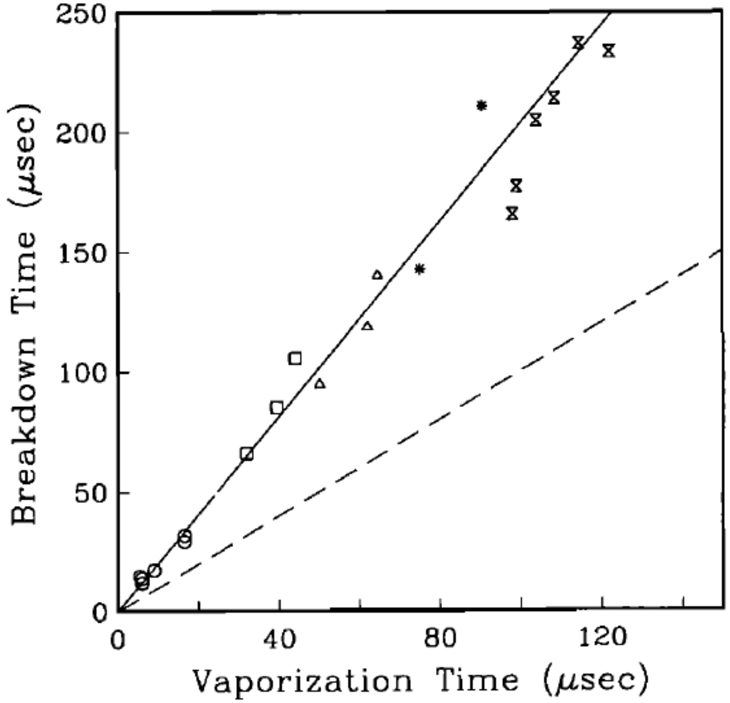
$$E_{\max} \cong 0.6/t_p^{0.33} A^{0.1} \quad (negative) \qquad E_{\max} \cong 0.3/t_p^{0.33} A^{0.1} \quad (positive).$$

Material	Relative Dielectric Constant ($\varepsilon/\varepsilon_0$)	Dielectric Strength (MV/cm)
Transformer oil	3.4	1
Mylar	3	1.8
Polyethylene	2.25	1.8
Teflon	2.1	4.3
Polycarbonate	2.96	5.5

Bubble mechanism is valid for a long pulse (> 1us)

- For large pulse widths (i.e. several microseconds to dc), especially in high conductive water solutions, the process of breakdown is preceded by vapor formation due to heating by the pre-breakdown current in the liquid.

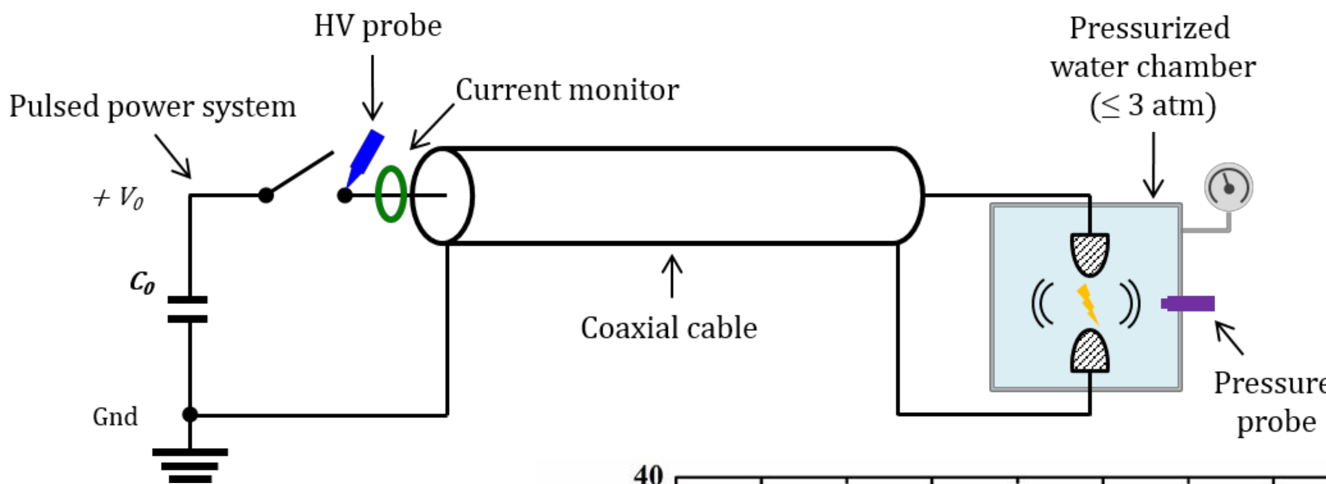




- However, bubble formation before plasma formation in the case of sub-us pulses is likely to be incomplete and transient as bubble formation is typically at a time scale of at least 1μs.

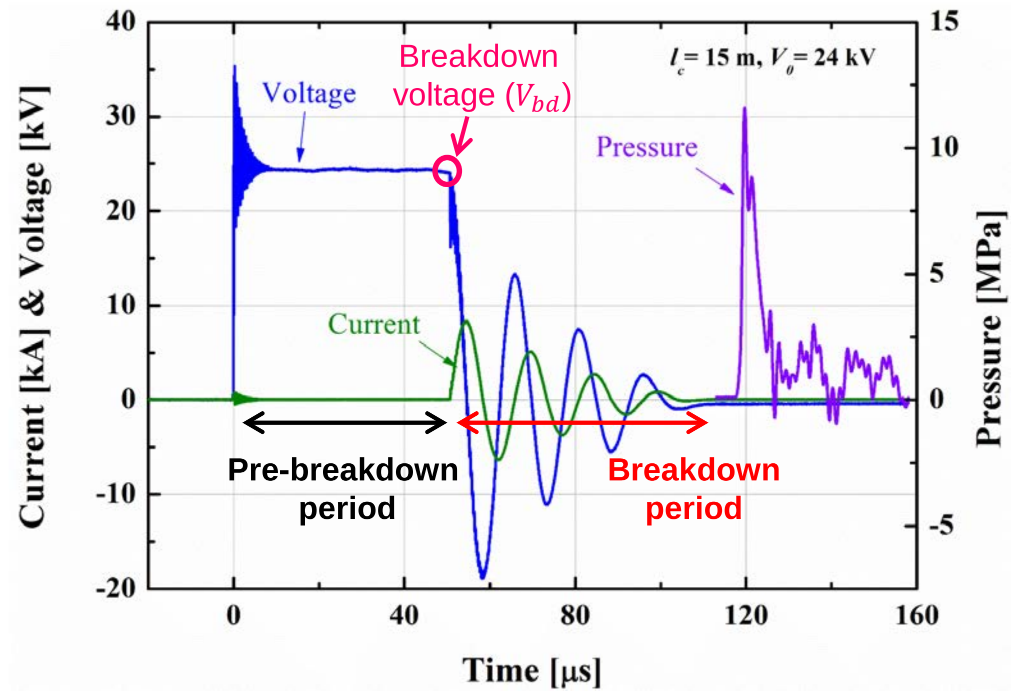
A. Olsen, J. Acoust. Soc. Am. 94, 2226 (1993)

Typical characteristics of electrical breakdown of water



Applications

- Oil well stimulation
- Rock fragmentation
- Material Processing
- Ultrasound acoustic source
- Well screen cleaning
- Green algae treatment



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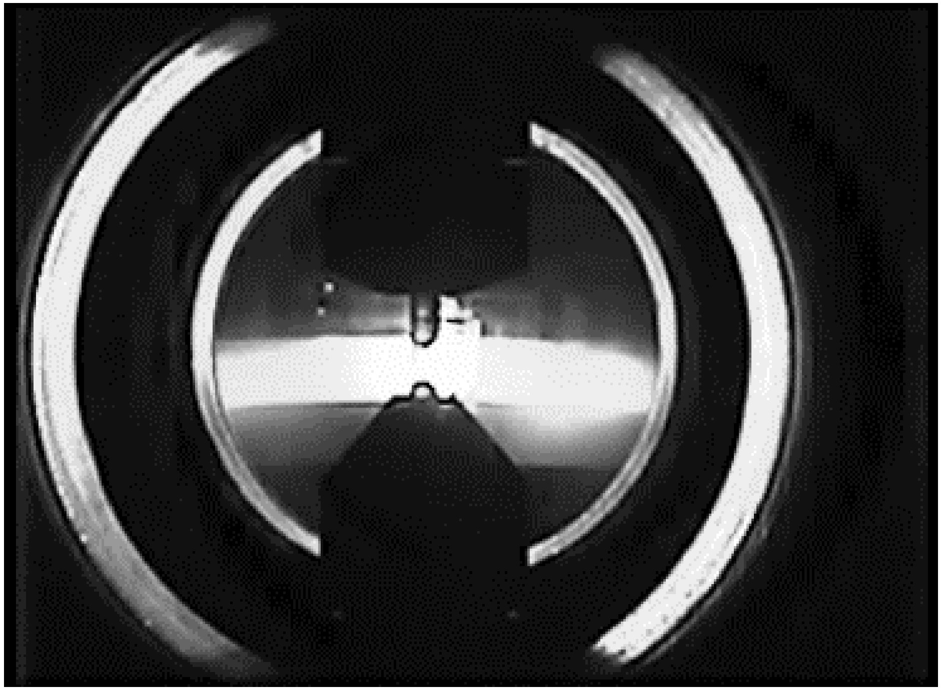
Fast framing movie

Water breakdown

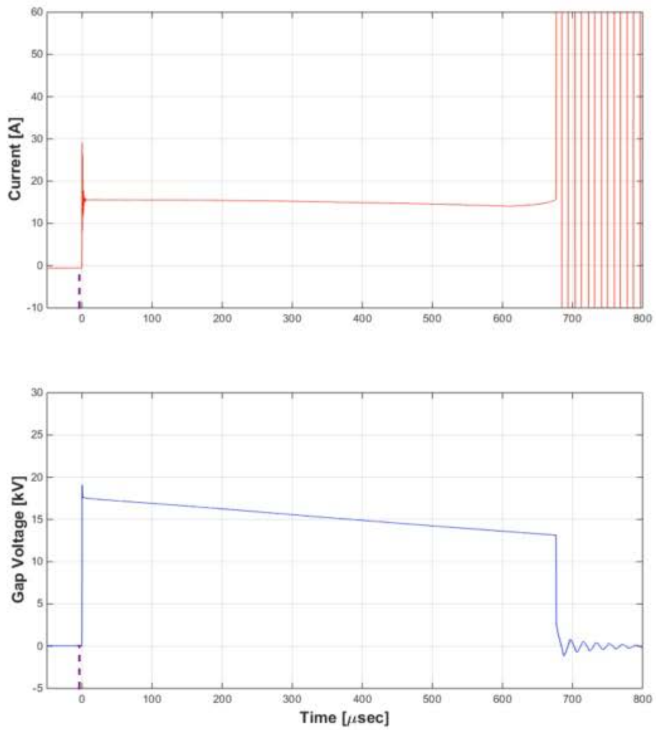
Bubble implosion

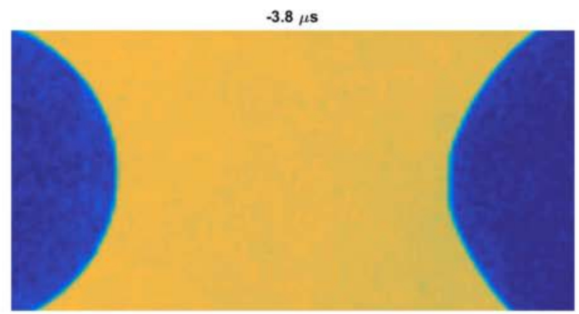


1 sec (playing time) = 0.1 ms (real time)

Water breakdown process (positive mode)

$V_0 = 16\ kV$

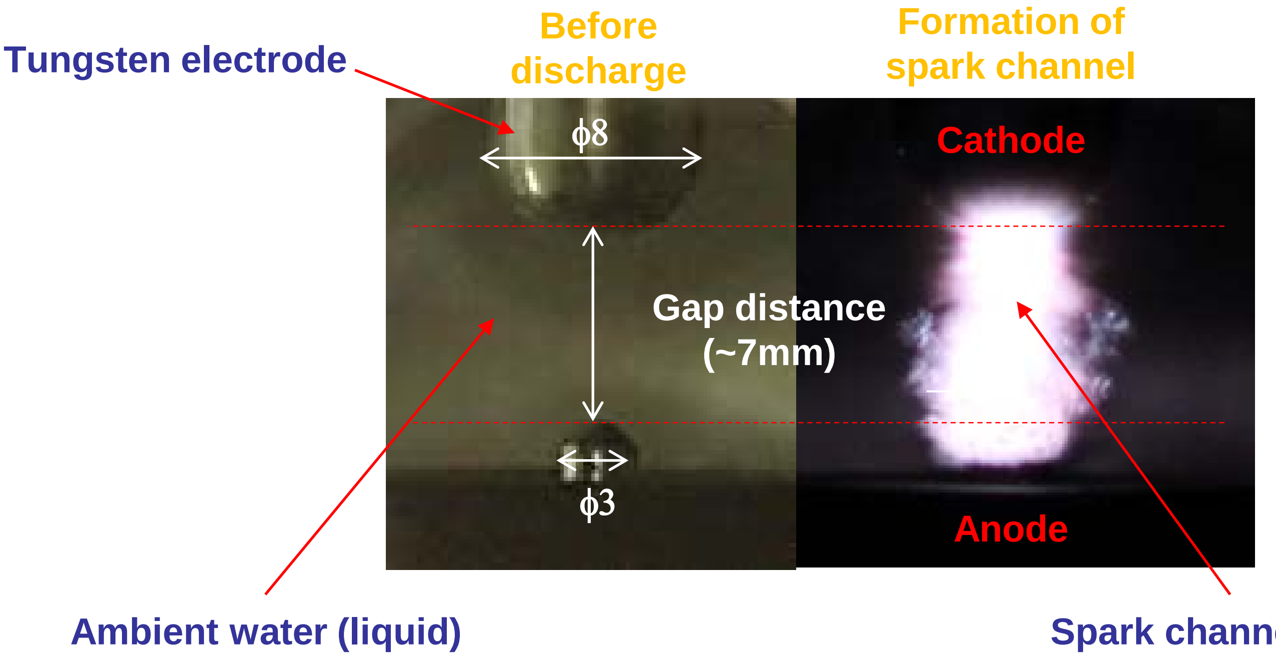


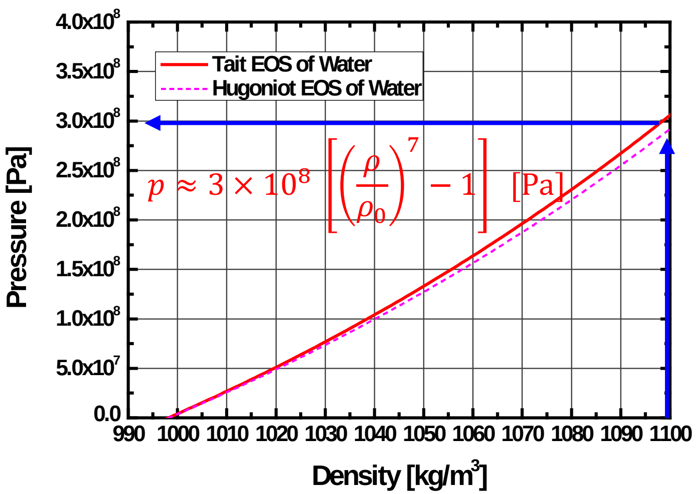


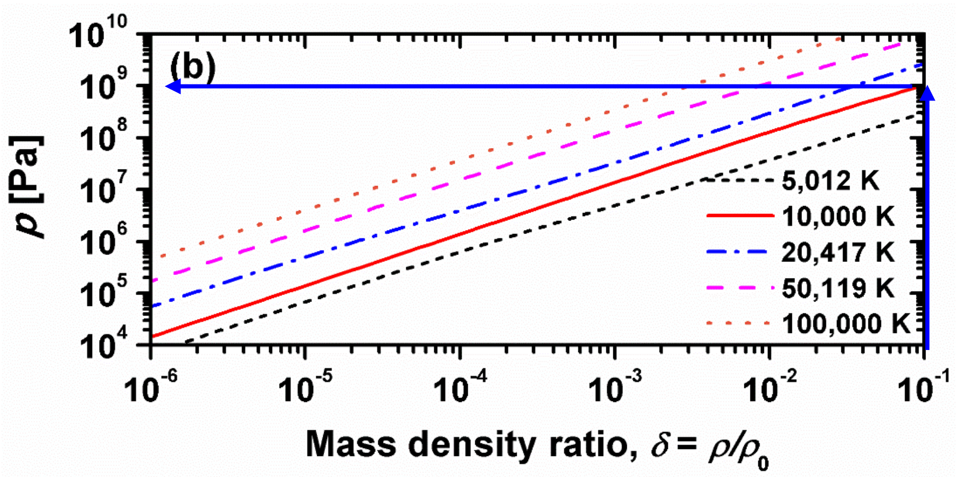
cathode

anode

Generation of strong shockwaves in water





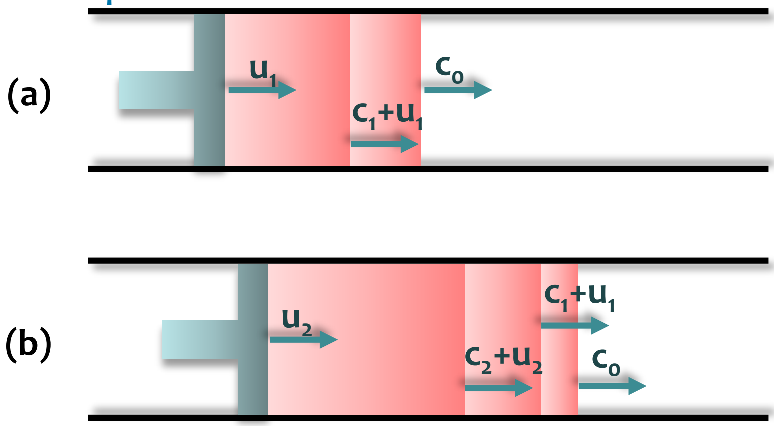


Mechanism for shock wave generation by pulsed spark discharge

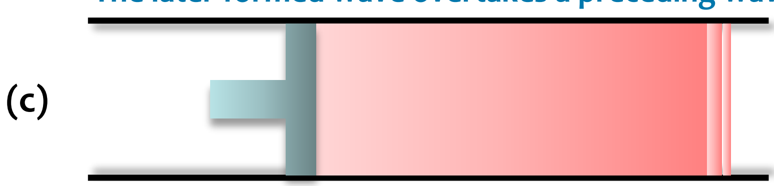
Diagram of the shock wave formation

First compression wave with speed of sound

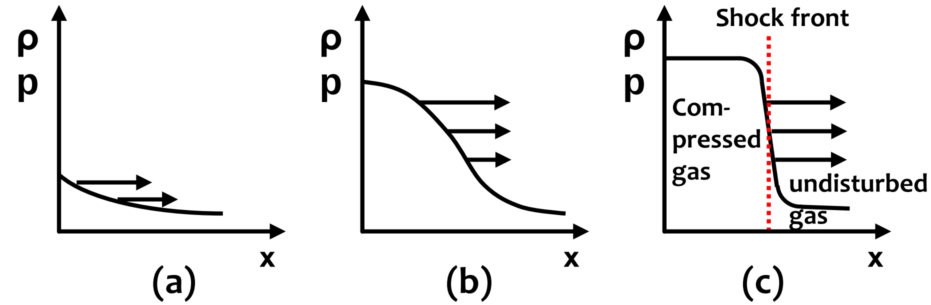
Second wave propagation with a higher speed than speed of sound

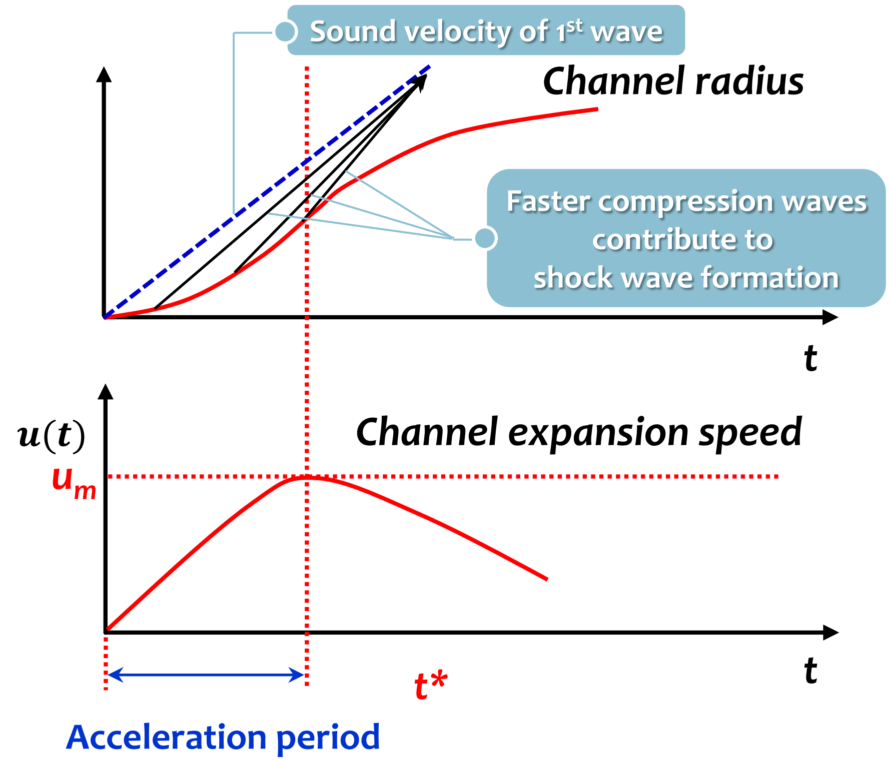


The later formed wave overtakes a preceding wave



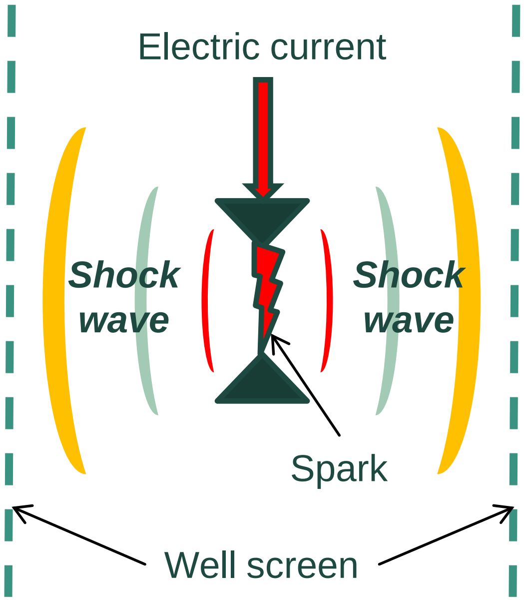
* $c_{0,1,2}$: sound velocity, $u_{1,2}$: particle velocity

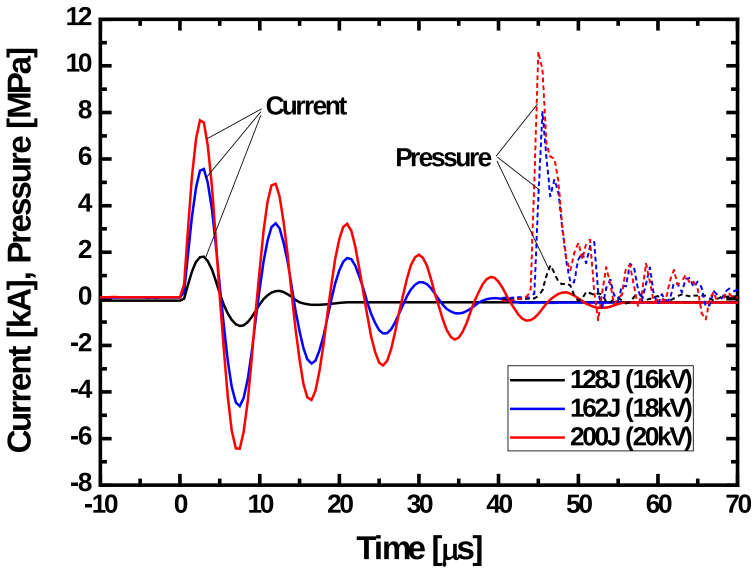


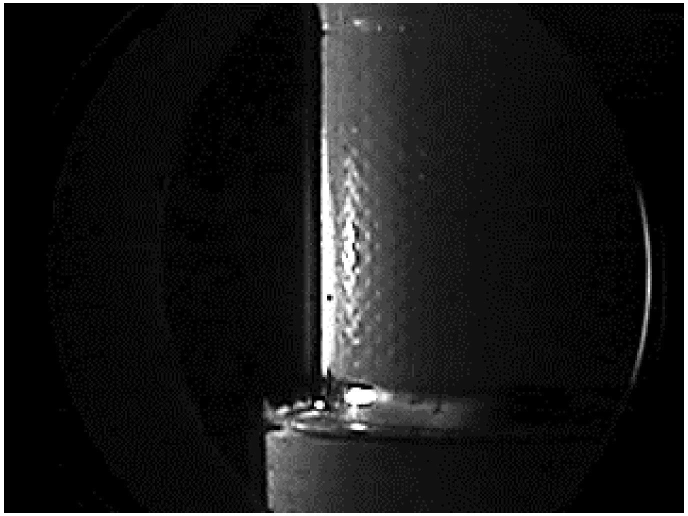


[5] Y. B. Zel'dovich and Y. P. Raizer, "*Physics of Shock Waves and High-Temperature Hydrodynamic Phenomena*", Dover Publications, New York (2002)

Application to water well cleaning



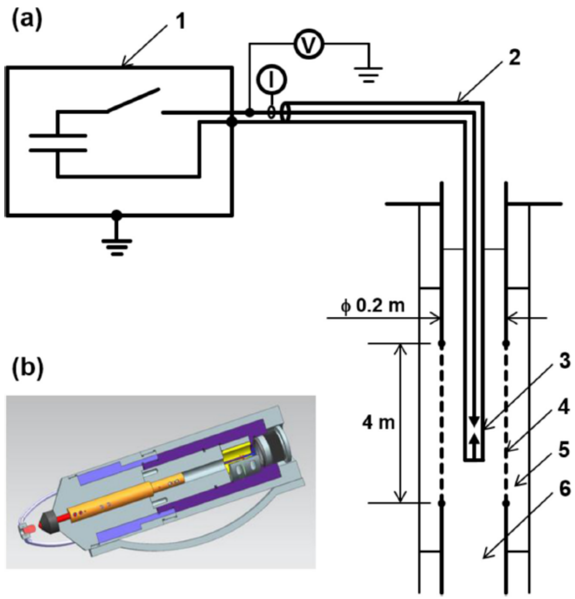


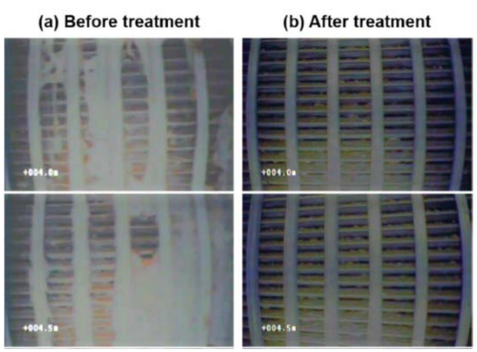


K. J. Chung et al., The Journal of Engineering Geology 23, 29 (2013) (Korean)

Application to water well cleaning

Vertical well cleaning



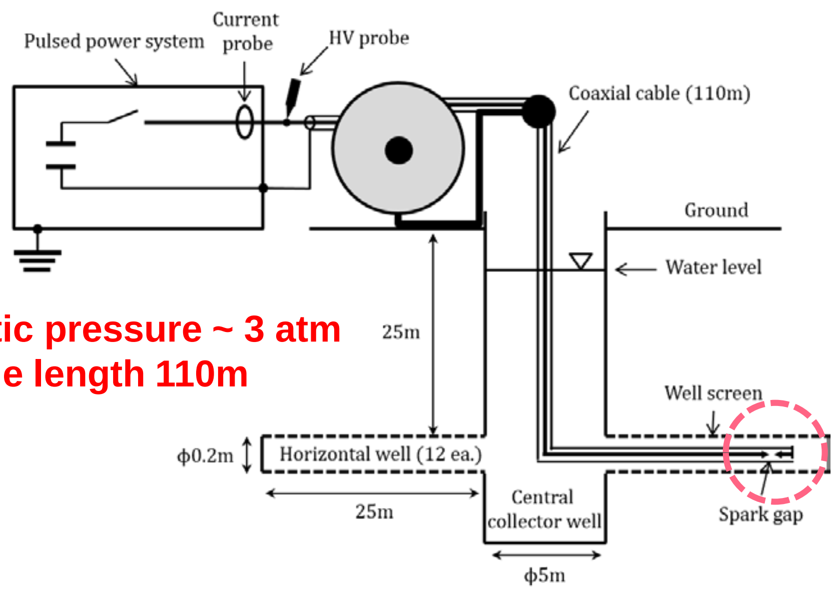


(1) Pulsed power system
(2) Coaxial cable (15m)
(3) Spark generator
(4) Well screen
(5) Gravel pack

Hydraulic conductivity (Hvorslev theory)

$$: 2.35 \times 10^{-4} \rightarrow 6.73 \times 10^{-4} \ [m/s]$$

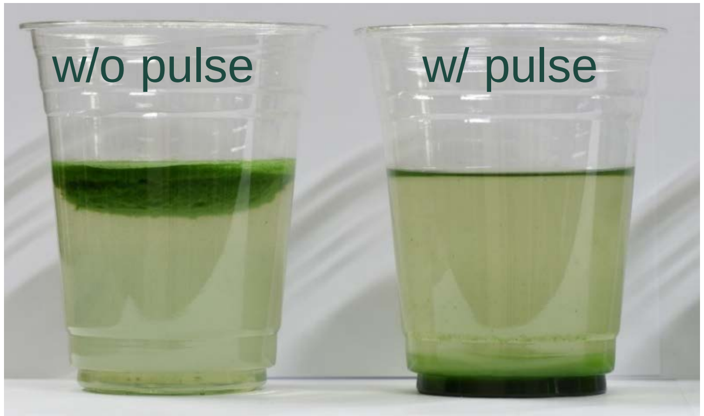
Horizontal well cleaning

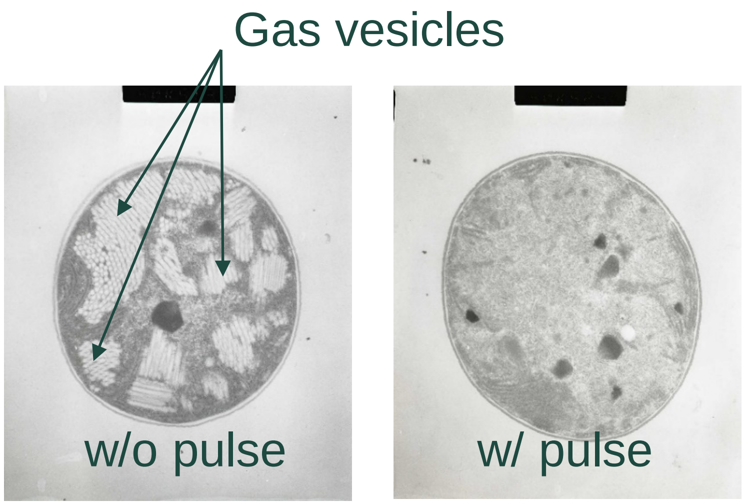


Hydrostatic pressure ~ 3 atm
Cable length 110m

Increased well efficiency ~ 21.3%

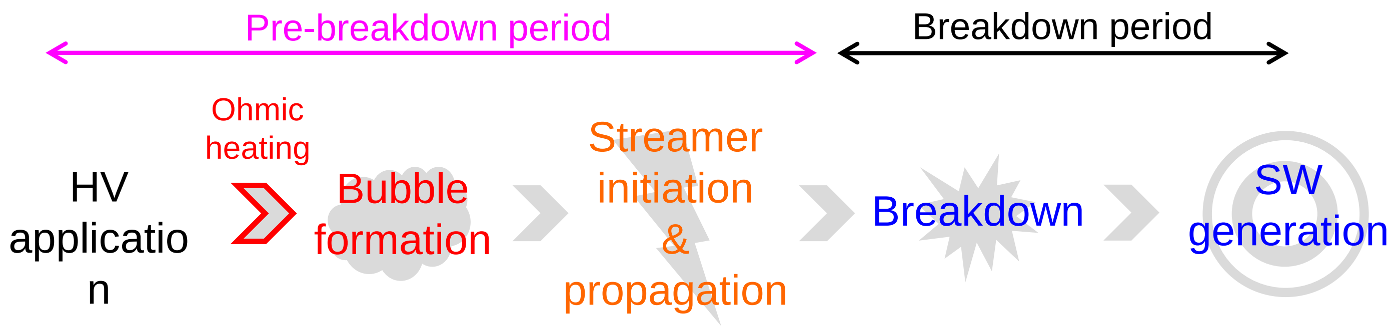
Application to green algae treatment





→ Shock wave destroys gas vesicles to sink the water-bloom down to the bottom

Key features of subsonic discharge



Key features

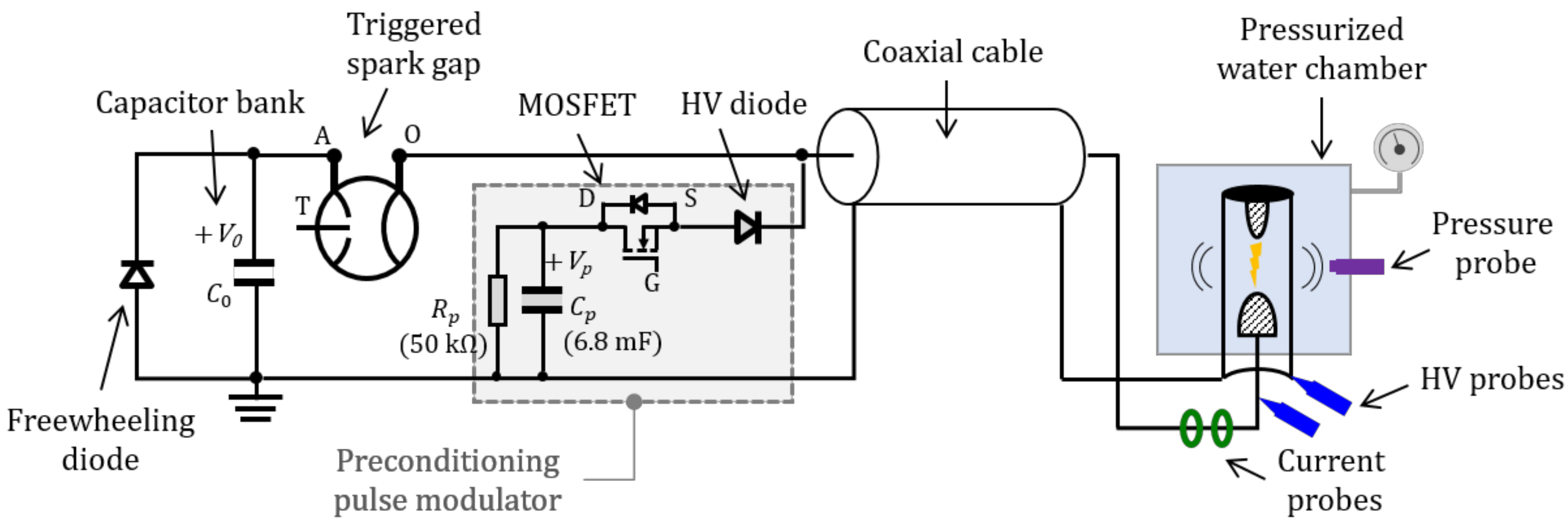
- Input energy ~ a few hundred J/pulse to generate spark discharges
- High breakdown strength of liquid
  - → Discharge initiation from the low density region (i.e. bubble or vapor layer)
  - → Pre-existing bubbles are particularly important

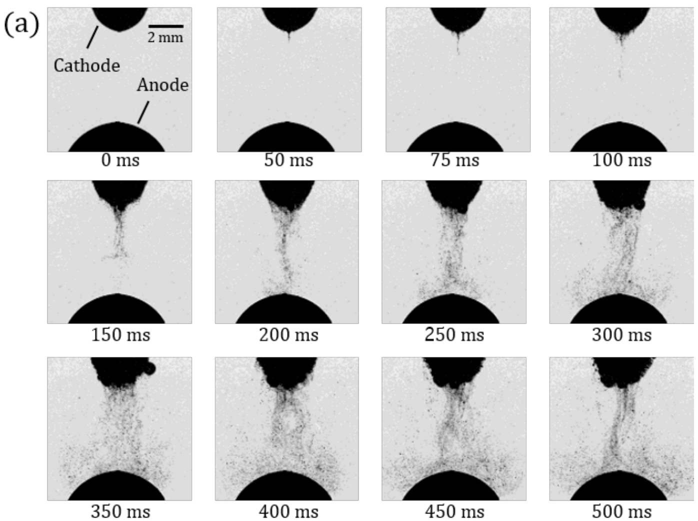
Circuit response during the pre-breakdown period ⇒ $\boldsymbol{V_{bd}}$

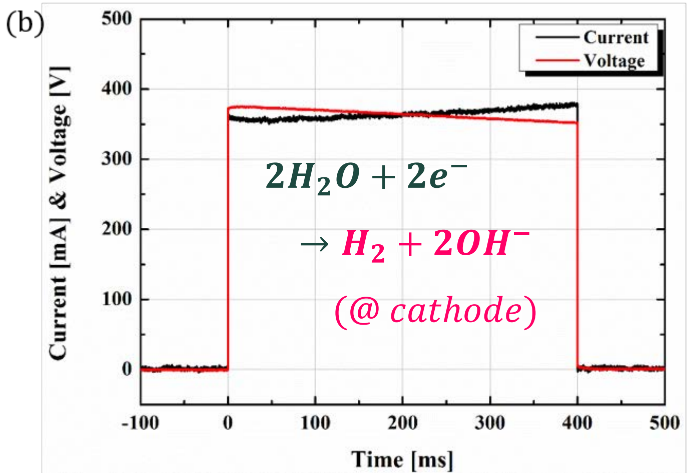
& Thermo-electric features of underwater streamer ⇒ $\boldsymbol{r_0, \rho_0, T_0, R_0}$

→ **Crucial in SW generation**

Use of water electrolysis for pre-breakdown acceleration

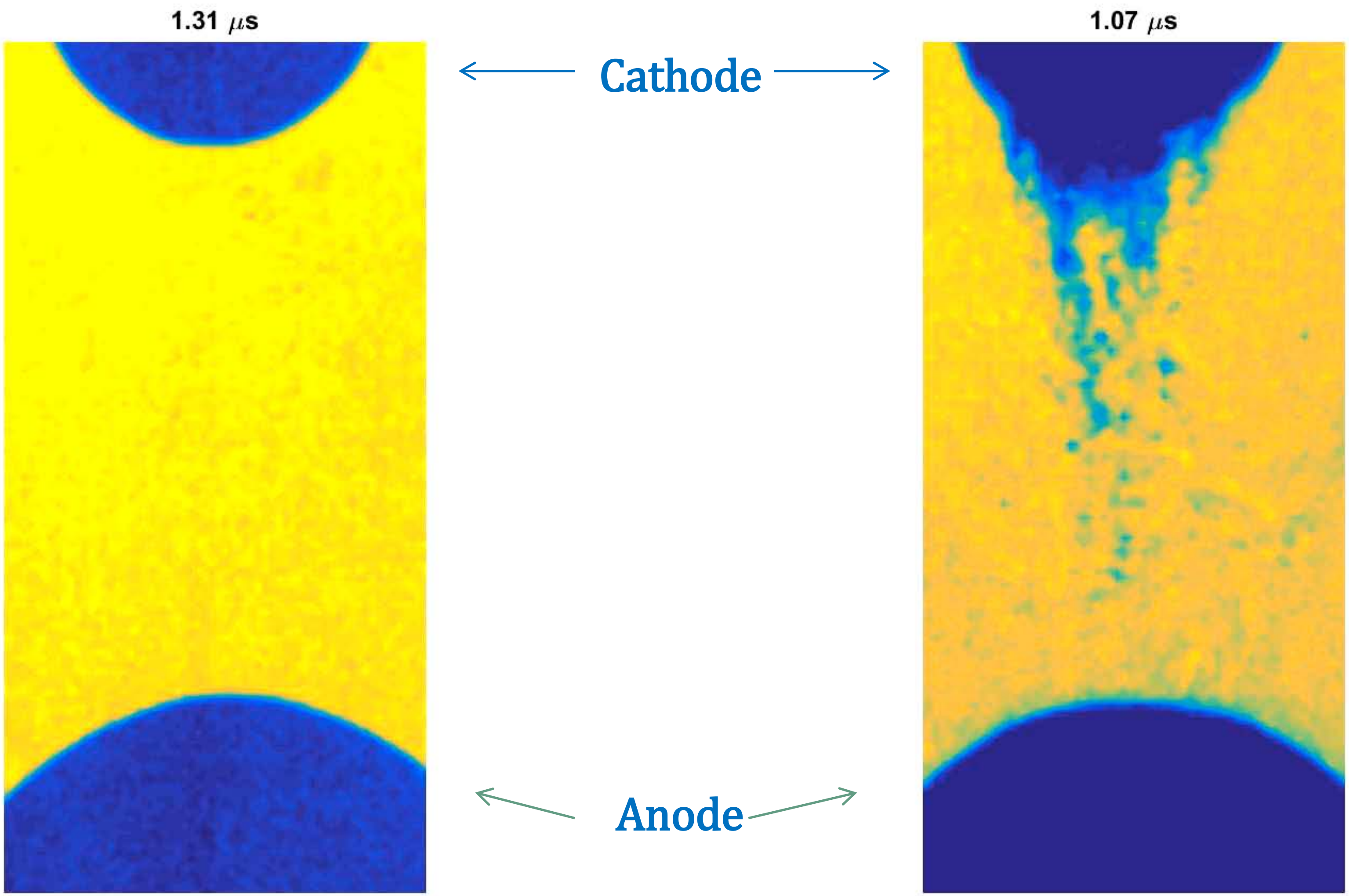






K. Lee *et al*, Appl. Phys. Lett. **112** (2018) 134101

Pre-breakdown acceleration by water electrolysis



Negative streamer discharge

- Conventional positive mode

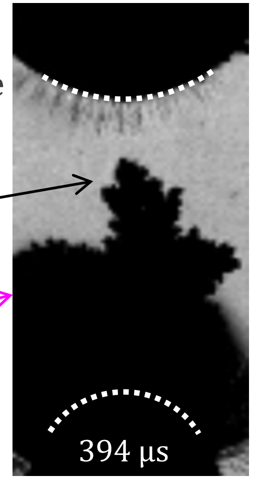
Cathode

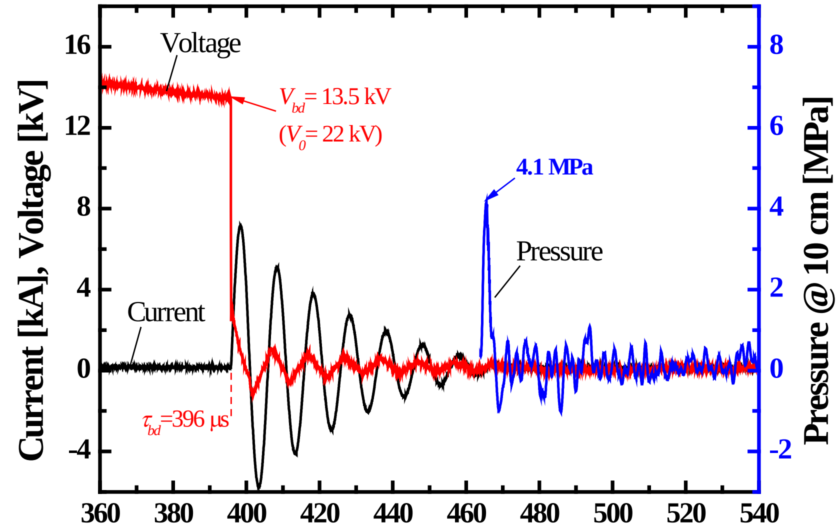
Positive streamer

Bubble cluster

Large uncertainty

Anode





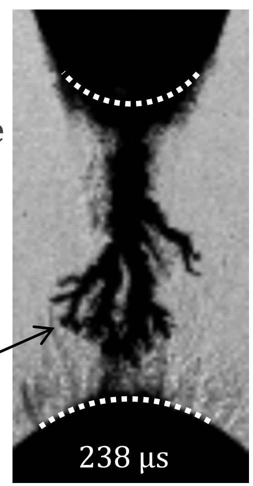
- Accelerated negative mode

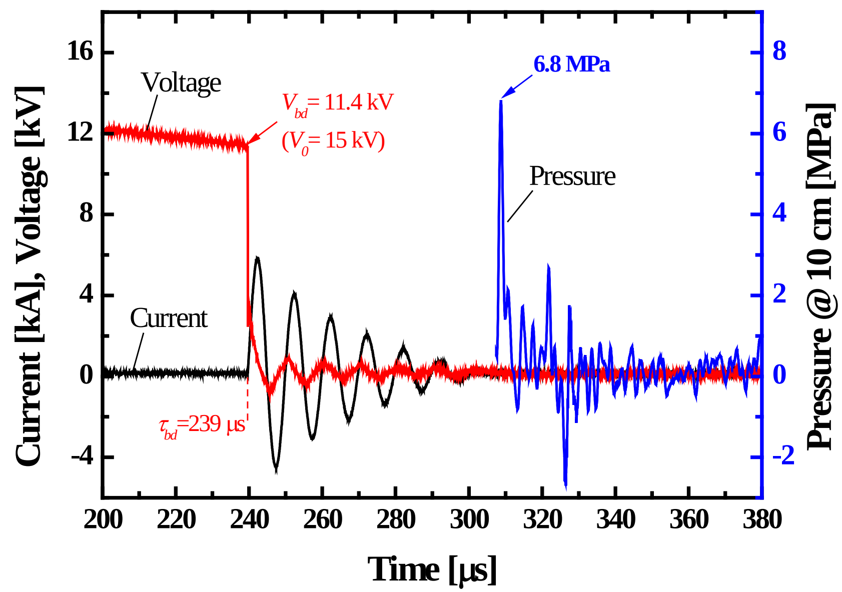
Cathode

More suitable to investigate the mechanism of shock wave development

Negative streamer

Anode





Streamer propagation