Dielectric fluid in inhomogeneous pulsed electric fields

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Workshop On Non-Equilibrium Flow Phenomena
In Honor of Mikhail Ivanov’s 70\textsuperscript{th} Birthday

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Hornung et al. (1979) put forward a hypothesis that a hysteresis may exist in transition between RR and MR.

If the angle of incidence is changed continuously, the transition from $RR \rightarrow MR$ and $MR \rightarrow RR$ transition may occur at different values of the incidence angle.

$RR \rightarrow MR$ transition occurs at $\theta = \theta_{LO}$, the reverse $MR \rightarrow RR$ transition occurs at $\theta = \theta_{H}$. 

$\theta = 24.0^\circ$
Motivation

• New experimental capabilities: High-voltage nanosecond power sources

• Interesting multidisciplinary physical problem

• Many potential applications:
  ➢ Volumetric “cold” ionization in liquid phase. Fuel modification
  ➢ Plasma medical treatment
  ➢ High voltage insulation
  ➢ High-current switching
  ➢ Cavitation study
Outline

- Nano- and Sub-Nanosecond breakdown in liquids
- Compressible fluid model with electrostrictive volumetric force
- Cavitation development in pulsed non-uniform electric field
- Characteristic areas in the vicinity of a needle-like electrode
- A single cavitation void in electric field
- Water microdroplet in oil
- Conclusions
Temporal dynamics of nanosecond discharge

Point-Plane
Electrode Diameter: 100 μm
Inter-electrode Distance: 3 mm


Nanosecond breakdown in liquid dielectric looks like gas breakdown

But, the nature is different and was explained in 2013
ns breakdown in water

The condition of breakdown: \((E / N) > (E / N)_{br}\), \(\frac{N_{\text{Water}}}{N_{\text{air}}} \approx 1000\)

The breakdown field in air at \(p=1 \text{ Atm}\) and \(T=300 \text{ K}\) is 30 kV/cm

Follow to the similarity of \(E/N\), condition of breakdown in water should be \(\sim 30 \text{ MV/cm}\)

But, in reality, it is in \(\sim 10\’s – 100\’s\) times smaller

The breakdown in liquids under long high-voltage pulse (\(\sim 0.1 – 100 \mu\text{s}\)) occurs in gas bubbles. in this case it is not very different from the usual breakdown in a gas, and has been well studied.

**Possible mechanisms of bubble creation in nanoseconds??**

1. **Thermal fluctuations**: \(4\pi r^2 \sigma \sim \frac{4}{3} \pi r^3 N_m kT\)

\(\sigma\) – surface tension coefficient: \(\sigma_{\text{water}} = 0.068 \text{ N/m}, \quad \sigma_{\text{PDMS}} = 0.020 \text{ N/m}\)

**Upper limit**: \(r \approx \frac{3\sigma}{N_m kT} \sim 1 \text{ nm}\)

2. **Joule heating**

Ultra pure water: \(\sigma=5\cdot10^{-6} \text{ S/m}\)

Regular tap water: \(\sigma =0.005 - 0.05 \text{ S/m}\)

\(j = \sigma E; \quad E = \frac{V}{r} \cdot \frac{1}{\ln d/r}\)

\(W_j = \sigma E^2 \cdot \tau \sim \sigma \left(\frac{V}{r_0}\right)^2 \cdot \tau\)

\(\tau = 10^{-9} \text{ s}\)

\(W_j = \left(10^6 - 10^8\right) \text{ J/m}^3\)

**Evaporation**

\(\kappa = 2257 \text{ kJ/kg}\)

\(W_{\text{evap}} = \kappa \cdot \rho \approx 2.3 \cdot 10^9 \text{ J/m}^3\)

**Expansion Work (pV)**

\(W_{pV} \approx 10^5 \text{ J/m}^3\)

at \(p = 1 \text{ Atm}\)

For pulse duration \(\tau \sim 1 \text{ ns}\), \(W_J < \ll W_{\text{evap}} + W_{pV}\)

Gas bubble creation in nanoseconds is impossible!
A bubble-free breakdown mechanism at nanosecond high-voltage pulses

Shneider, Pekker, Fridman, IEEE Transactions on Dielectrics and Electrical Insulation (2013)

- It is known that at certain values of the stress tension (negative pressure) – cavitation occurs
- Such negative pressure arises faster than in ps-time scale in the vicinity of a needle electrode under ponderomotive electrostrictive action of highly non-uniform electric field
- A large amount of micro-pores forms in the vicinity of needle electrode where liquid becomes a highly dispersive media

\[
\vec{F} = e\delta n\vec{E} - \frac{\varepsilon_0}{2} E^2 \nabla \varepsilon + \frac{\varepsilon_0}{2} \nabla \left( \frac{E^2}{\rho} \right)
\]

For nonpolar dielectrics:
\[
\vec{F} = \frac{\varepsilon_0}{2} \nabla \left( \frac{E^2}{\rho} \right) = \frac{\varepsilon_0}{2} (\varepsilon - 1)(\varepsilon + 2) \nabla E^2
\]

For polar dielectrics:
\[
\vec{F} = \frac{\varepsilon_0}{2} \nabla \left( \frac{E^2}{\rho} \right) \approx 0.5 \varepsilon_0 \varepsilon \nabla E^2 \quad (\alpha \approx 1.5)
\]

If the conditions for cavitation are satisfied, pores can appear in any point where \( P < P_c = -(5-100) \) MPa.

\[
P_{tot} = -\left( 0.5 \varepsilon_0 E^2 \frac{\partial \varepsilon}{\partial \rho} \rho - p \right); \quad |P_E| = 0.5 \varepsilon_0 E^2 \frac{\partial \varepsilon}{\partial \rho} \rho \approx 0.5 \alpha \varepsilon_0 \varepsilon E^2 \approx \varepsilon_0 \varepsilon \frac{U^2 r_0^2}{R^4} > |P_c|
\]

\[
R < r_0 \left( \varepsilon_0 \frac{U^2}{|P_c| r_0^2} \right)^{1/4}
\]

The region with cavitation, \( R(V,r_0) \) can be measured in experiments!

Electric field inside a spherical micropore
\[
E_p = \frac{3\varepsilon}{1 + 2\varepsilon} E
\]

Ionization condition (necessary for breakdown): \( \Sigma \approx 2E_p r_p \geq I \sim 10 \text{ eV} \)
**Cavitation**

Cavitation is the formation of voids or gas bubbles in a liquid in a region where the pressure in the liquid falls below its vapor pressure.

Usually, cavitation is observed in the flow regions where pressure is negative: 1. Centrifugation; 2. Behind shock waves; 3. The reverberation phase of the acoustic wave, etc.

Depends on the purity of liquid; temperature; duration of the negative pressure phase

Measurements of the critical parameters for cavitation development is a serious issue for many applications; **huge uncertainty of data**

**HERBERT, BALIBAR, AND CAUPIN**

**PHYSICAL REVIEW E 74, 041603 (2006)**

**TABLE I.** Comparison between different cavitation experiments. Among the numerous and scattered values of the cavitation pressure in the literature, only the most negative have been selected.

<table>
<thead>
<tr>
<th>Method</th>
<th>Ref.</th>
<th>$T$ (°C)</th>
<th>$V$ (mm$^3$)</th>
<th>$\tau$ (s)</th>
<th>$J = 1/(V\tau)$ (mm$^{-3}$ s$^{-1}$)</th>
<th>Wall</th>
<th>$P_{cav}$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Berthelot</td>
<td>[17]</td>
<td>40</td>
<td>1</td>
<td>20</td>
<td>$5 \times 10^{-2}$</td>
<td>Pyrex glass</td>
<td>$-16$</td>
</tr>
<tr>
<td>Berthelot</td>
<td>[19]</td>
<td>53</td>
<td>47</td>
<td>5</td>
<td>$4.3 \times 10^{-3}$</td>
<td>Stainless steel</td>
<td>$-18.5$</td>
</tr>
<tr>
<td>Centrifugation</td>
<td>[21]</td>
<td>10</td>
<td>0.38</td>
<td>10</td>
<td>$2.6 \times 10^{-1}$</td>
<td>Pyrex glass</td>
<td>$-27.7$</td>
</tr>
<tr>
<td>Shock wave</td>
<td>[22]</td>
<td>25</td>
<td>0.003</td>
<td>$10^{-8}$</td>
<td>$3.3 \times 10^{10}$</td>
<td>Silica fiber</td>
<td>$-27$</td>
</tr>
<tr>
<td>Acoustic</td>
<td>[26]</td>
<td>30</td>
<td>200</td>
<td>0.1</td>
<td>$5 \times 10^{-2}$</td>
<td>None</td>
<td>$-21$</td>
</tr>
<tr>
<td>Inclusions</td>
<td>[30]</td>
<td>40–47</td>
<td>$4.2 \times 10^{-6}$</td>
<td>1</td>
<td>$2.4 \times 10^{5}$</td>
<td>Quartz</td>
<td>$-140$</td>
</tr>
<tr>
<td>Acoustic</td>
<td>this work</td>
<td>20</td>
<td>$2.1 \times 10^{-4}$</td>
<td>$4.5 \times 10^{-8}$</td>
<td>$1.1 \times 10^{11}$</td>
<td>None</td>
<td>$-24$</td>
</tr>
</tbody>
</table>

Combination of laser scattering and nanosecond high-voltage pulses in liquids may serve for the cavitation parameters definition
Cavitation

- Propeller damage
- Noise
- Engine low efficiency
- Enhanced fuel consumption

Very important to know critical flow parameters!
Observation of cavitation

A comparison of the experimentally observed dimensions of the region where cavitation develops in the controlled unsteady conditions with the results of calculations in the framework of the hydrodynamic model allows to obtain the critical parameters of cavitation initiation: $P_{cr}$, $J$.


![Diagram](image)

**Figure 1.** Optical observation of the cavitation region. The radius of curvature of the electrode is $R_0 = 35 \, \mu m$. The distance between the needle tip and the flat electrode is $d_p = 1.5 \, mm$. (a) The voltage between the electrodes is zero. (b) The voltage between the electrodes is maximum.


<table>
<thead>
<tr>
<th>Voltage</th>
<th>0 ns</th>
<th>1 ns</th>
<th>2 ns</th>
<th>3 ns</th>
<th>4 ns</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.6kV</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9.72kV</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Size of the cavitation area agrees well with the predictions of a theory.

No cavitation. The same in a theory.
Macroscopic fluid model of induced flow

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{u}) = 0,
\]

\[
\rho \left( \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla)\vec{u} \right) = -\nabla p + \tilde{F} + \eta_d \left[ \Delta \vec{u} + \frac{1}{3} \nabla (\nabla \cdot \vec{u}) \right]
\]

and the Tait equation of state for “compressible” water:

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

\[
\tilde{F} = \frac{\varepsilon_0}{2} \nabla \left( E^2 \frac{\partial \varepsilon}{\partial \rho} \rho \right) \approx 0.5 \alpha \varepsilon_0 \varepsilon \nabla E^2 \quad (\alpha \approx 1.5)
\]

\[
\rho_0 = 1000 \text{ kg/m}^3
\]

\[
p_0 = 10^5 \text{ Pa}
\]

\[
B = 3.07 \cdot 10^8 \text{ Pa}
\]

\[
\gamma = 7.5
\]

\[
U(t) = U_0 t / t_0, \quad t \leq t_0
\]

\[
t_0 - \text{pulse front}
\]

The area of integration and the grid in prolate spheroidal coordinates

Macroscopic fluid model of induced flow

\[ U(t) = \frac{U_0 t}{t_0}, t \leq t_0 \]

\[ U_0 = 7.5 \text{ kV}; \quad r_{el} = 5 \text{ um} \]

Contours of the total and ponderomotive pressures, and the relative density perturbation at \( t = 5 \text{ ns} \) for a pulse with the front duration \( t_0 = 5 \text{ ns} \)

Macroscopic fluid model: pulse front duration $t_0$ matters!

$U(t) = U_0 t / t_0, t \leq t_0$

$U_0 = 7.5 \text{ kV}; r_{el} = 5 \text{ um}$

$p_E = -0.5 \alpha \varepsilon_0 \varepsilon E^2 \ (\alpha \approx 1.5)$

$p_{cav} = -30 \text{ MPa}$

Longitudinal distributions of the total pressure $p_{tot} = p + p_E$; flow velocity and relative density along the symmetry axis ($r=0, z$). Curve 1 corresponds to the $t/t_0 = 0.25, 2 - 0.5, 3 - 0.75, 4 - 1$. The dashed line shows the pressure threshold for cavitation

Formation of negative pressure near the electrode after a sharp shutdown of the voltage pulse

As $t=0$, the hydrostatic pressure was taken equal to the absolute value of electrostrictive $p \approx |p_E| = 0.5\varepsilon\varepsilon_0 E^2$ at the maximum voltage on the electrode $U_0$. 

$\rho_E \gg \rho_0, p$  $p_E + p \sim \rho_0$,  $p \gg \rho_E, \rho_0$
Cavitation development

Follow Zel’dovich (1942) and Fisher (1948) theory for liquid under negative pressure at \( E=0 \).

The energy required to create a bubble of the radius \( R \) in the liquid is equal:

\[
\Sigma(R) = \frac{4}{3} \pi R^3 (P - P_{sat}) + 4\pi R^2 \sigma
\]

The critical radius of the bubble, after which it will expand, and the work for its creation:

\[
R_{cr} = \frac{2\sigma}{P_{sat} - P}, \quad \Sigma_{cr} = \frac{16\pi \sigma^3}{3(P_{sat} - P)^2}
\]

The rate of generation of cavitation voids of critical size \([1/m^3s]\):

\[
\Gamma = \frac{3k_B T}{16\pi (\sigma \cdot k_\sigma)^3} \frac{P^3}{4\pi \hbar} \exp \left( -\frac{16\pi (\sigma \cdot k_\sigma)^3}{3k_B T \cdot P^2} \right)
\]

Modification of Zel’dovich-Fisher theory for liquid under the electrostrictive negative pressure with taking into account the electric field perturbation (Shneider, Pekker J. Appl. Phys (2013))

\[
\vec{F} = -\frac{\varepsilon_0}{2} E^2 \nabla \varepsilon + \frac{\varepsilon_0}{2} \nabla \left( E^2 \frac{\partial \varepsilon}{\partial \rho} \rho \right)
\]

The energy required to create the pore of the radius \( R \) (at \( \varepsilon >>1 \)):

\[
\Sigma(R) = 4\pi R^2 \sigma - \frac{5\pi R^2}{2} \left( \alpha - 1 \right) \frac{\varepsilon_0 E^2}{2} = 4\pi R^2 \sigma + \frac{5\pi R^2}{2} \bar{P} \quad \bar{P} = -\frac{1}{2} (\alpha - 1) \varepsilon_0 \varepsilon E^2
\]

The critical radius and the energy required to create the cavitation pore are:

\[
R_{cr} = \frac{16}{15} \frac{\sigma}{\bar{P}}, \quad \Sigma_{cr} = \pi \frac{16^2}{15^2} \frac{4 \sigma^3}{3 \bar{P}^2} \approx 1.517\pi \frac{\sigma^3}{\bar{P}^2}
\]

The effective critical electrostrictive pressure: \( \bar{P}_{cr} \approx (\alpha - 1) P_{cr} / \alpha \approx 0.33 P_{cr} \); \( P_{cr} = -0.5 \varepsilon_0 \varepsilon E_0^2 \approx -30 \text{ MPa} \)
Cavitation development: example

Voltage pulse: \( U = U_0 t / t_0 \); \( U_0 = 22.4 \) kV ; \( t_0 = 3 \) ns;  Spherical electrode, \( r_0 = 50 \) \( \mu \)m

At \( P_{cr} = -30 \) Mpa; \( R_{cr} \approx 1.48 \) nm

The rate of generation of cavitation voids of critical radius \( R_{cr} \left( P_E (r, t) \right) \), normalized by \( \Gamma_0 = \Gamma \left( \text{at} \ P = -30.6 \text{ MPa} \right) \approx 5.7 \cdot 10^{24} \) m\(^{-3}\)s\(^{-1}\).
Development of micropores in a dielectric liquid in inhomogeneous pulsed electric fields

\[ \vec{F} = -\frac{\varepsilon_0}{2} E^2 \nabla \varepsilon + \frac{\varepsilon_0}{2} \nabla \left( E^2 \frac{\partial \varepsilon}{\partial \rho} \rho \right) \]

The surface forces exerted by the electric field on the surface of the pore per unit area

\[ F_{S,n} = \frac{\varepsilon_0}{2} \left( \alpha \left( \frac{E^2_{p,n}}{\varepsilon} + \varepsilon E^2_{p,t} \right) - (\varepsilon - 1) \left( \frac{E^2_{p,t}}{\varepsilon} + \frac{E^2_{p,n}}{\varepsilon} \right) \right) - \frac{2\sigma}{R} - p \quad (*) \]

The mean pressure acting on the micropore surface, averaging (*) over the sphere surface \(4\pi R^2\),

\[ P_{av} = (3/4)(\alpha - 1)\varepsilon_0 \varepsilon E^2 - 2\sigma / R - p \quad (**) \]

From the continuity equation for incompressible fluid:

\[ u(r',t) = \frac{R^2}{r'^2} U, \quad U = dR / dt; \quad r' > R \]

The kinetic energy of the fluid is:

\[ W_K = \frac{1}{2} \int_{r=R}^{\infty} 4\pi \rho u^2 r'^2 dr' = 2\pi \rho R^3 \left( \frac{dR}{dt} \right)^2 \]

The relevant work of the pressure forces (**) for the pores expansion is:

\[ W_p = 4\pi \int_{R_0}^{R} P_{av} r^2 dr \]

From the condition: \( W_K = W_p \), the equation for dynamic of the pore radius

\[ \frac{d}{dt} \left( R^3 \left( \frac{dR}{dt} \right)^2 \right) = \frac{4}{\rho} \frac{dR}{dt} \left( \frac{3}{8} R (\alpha - 1) \varepsilon_0 \varepsilon E^2 - \sigma \right) \]

Characteristic regions in the vicinity of the electrode

**Region 1:** The electric field gradient is greatest; the occurring cavitation nanopores have enough time during the nanosecond voltage pulse to grow to a size at which an electron can gain enough energy for the excitation and the ionization of molecules of the liquid on the pore wall.

**Region 2:** The electrostrictive negative pressure reaches values at which the cavitation development becomes possible (which can be registered by the optical methods), but nanovoids appearing during the voltage pulse do not have enough time to grow to the size at which the potential difference across their borders becomes sufficient for the ionization or the excitation of water molecules.

**Region 3:** The development of cavitation is impossible, since the spontaneously occurring nanovoids do not grow, because the value of the electrostrictive negative pressure cannot compete with the forces of surface tension.

Development of micropores in a dielectric liquid in inhomogeneous pulsed electric fields

Assumed parameters: spherical electrode $r_0 = 100$ µm, $U_0 = 54$ kV, $t_0 = 3$ ns; water

Dependencies of $P_E$ (a) and the pore radius $R$ (b) at the time moment $t = t_0 = 3$ ns

Parameter $\Delta\phi = 2E_{in} R$ at the time moment $t = t_0 = 3$ ns

The solid vertical line corresponds to $\Delta\phi = 10$ eV

$E_{in} = \frac{3\varepsilon}{1 + 2\varepsilon} E \approx 1.5E$

The volumetric force distribution and the shape of the expanding micropores in water

Unperturbed E-field is quasiuniform in the vicinity of the pore

\[ \varphi_{in} = -\frac{3\varepsilon}{\varepsilon_{in} + 2\varepsilon} E_0 r \cos(\theta), \quad r \leq R \]

\[ \varphi_{out} = -E_0 r \cos(\theta) \left( 1 + \frac{(\varepsilon - \varepsilon_{in}) R^3}{2\varepsilon + \varepsilon_{in} r^3} \right), \quad r > R \]

Landau, Lifshitz, Electrodynamics of Continuous Media, 1984

Pore or gas bubble in water: \( \varepsilon=81, \varepsilon_{in}=1 \)

\[ F(r', \theta)_{out} \approx \frac{3}{2} \alpha \varepsilon_0 E_0^2 \frac{(\varepsilon - 1)}{2 \varepsilon + 1} \frac{R^3}{r^4} \left( 5 \cos^2(\theta) - 1 \right) - 2 \left( 3 \cos^2(\theta) + 1 \right) \frac{R^3}{r^3} \frac{(\varepsilon - 1)}{2 \varepsilon + 1} \]

\[ F_0 = \frac{3}{2} \alpha \varepsilon_0 \frac{(\varepsilon - 1)}{2 \varepsilon + 1} \frac{E_0^2}{R} \left( 1 + 2 \frac{(\varepsilon - 1)}{2 \varepsilon + 1} \right) \]

Because of stretching, the relative volume changing is small.
Example: if pore is an extended ellipsoid \( a \sim 10 \text{ nm}, b \sim 1 \text{ nm} \).
At the pore density \( n_p \sim 10^3 \text{ 1/\mu m}^3 \), \( \Delta V/V \sim n_p a b^2 \sim 10^{-5} \)
Water microdroplet in transformer oil

Water microdroplet in oil: $\varepsilon = 2$, $\varepsilon_{in} = 81$

The corresponding electric field along the z axis, normalized to $E_0$.

The distribution of the electrostrictive pressure in transformer oil in the vicinity of water droplets (normalized to $|P_E| = \frac{1}{2} \alpha \varepsilon_0 \varepsilon E_0^2$).

When the electric field is turned on quickly, water microdroplets may initiate cavitation formation and breakdown in the transformer oil, i.e., determine its dielectric strength at essentially prebreakdown values of the electric field.

1. At the beginning, the strong inhomogeneous electric field in the vicinity of the needle electrode creates a region saturated by cavitation micropores.

2. In the pores, the primary electrons are accelerated by electric field to energies exceeding the potential of ionization of a molecule of water.

3. After neutralization of electrons at the electrode, positive charged liquid forms a virtual needle electrode, and electrostrictive conditions for the appearance of the next set of cavities are fulfilled.

4. Coulomb repulsion in non-quasineutral regions is an additional source for the negative pressure and cavitation enhancement.
Conclusions

• If the subnanosecond or nanosecond high-voltage pulses applied to the needlelike electrodes, the region of the negative pressure forms as a result of volumetric electrostrictive forces, which can lead to cavitation. (Similar effects can be stimulated by laser field)

• Analysis based on combination of nonstationary hydrodynamic calculations with laser scattering data in a real experiment may be a source of information on the cavitation threshold in liquid

• The cavitation micropores are strongly extended along the electric field and, even at their significant densities, relative change in volume is very small

• Water microdroplets may initiate cavitation and breakdown in transformer oil at essentially pre-breakdown values of the pulsed electric field

How the breakdown develops is a subject for further theoretical and experimental studies.
Thank you!

Questions?
Sonoluminescence at cavitation

Sonoluminescence is the emission of short bursts of light from imploding bubbles in a liquid when excited by sound.

Discovered by H. Frenzel and H. Schultes at the University of Cologne in 1934 as a result of work on sonar.

Bubble oscillations correspond to pressure anti-nodes.

From left to right: apparition of bubble, slow expansion, quick and sudden contraction, emission of light.

http://en.wikipedia.org/wiki/Sonoluminescence
Pulsed breakdown in high-voltage plain capacitor
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