

Formation mechanism of streamer discharges in liquids: a review

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Abstract: Streamer discharges in liquids have received lots of attention with respect to their considerable applications in various disciplines. Much effort has been spent to understand the basis of streamer ignition and propagation in dielectric liquids, but a comprehensive mechanism is far from conclusive. In this review, based on an introduction to the streamer physics and experimentally observed characteristics in various liquids, the authors emphasise on the current status of streamer discharge ignition mechanisms and present their understanding for each theory.

1 Introduction

When a high voltage is applied to an ionisable matter, filamentary discharges, so-called 'streamers' often appear at the first stage, in gases, liquids and solids. Streamers can propagate to non-ionised areas due to extensive electric field enhancement at their tips and the conducting channels in its interior [1–4]. The concept of streamer was first proposed by Raether [5], Loeb and Meek [6, 7], in 1930s, to explain the experimentally observed short breakdown time in high pressure gases, while the classical Townsend theory fails due to the neglect of space-charge effects induced by the electron avalanche. In the last decades, streamer discharge in liquids attracts increasing attentions due to its wide potential interest of applications, such as biomedicine [8, 9], purification [10–12] and decontamination [13, 14], high-voltage devices (e.g. power transformers, high-voltage insulation) [15–19], nanoscience [20] and food processing [21].

The mechanism of streamer-like breakdown in gas phase is in general well understood that direct impact ionisation dominates during the breakdown and other processes, such as natural background ionisation (e.g. generated by radiation, cosmic ray) [22, 23], photoionisation [24–27], Penning ionisation [28–30] or residual charges in pulsed discharges [31, 32] supply source electrons. However, streamer discharge in liquids is much less clear than in gases. A lot of effort has been spent to understand the underlying physics of streamers in various liquids and many reviews have been attempted [15, 33–38]. 'Direct impact ionisation' and 'gas bubble' are two classical mechanisms that has been proposed for almost a century [17] and are being frequently used currently to explain some experimental pre-breakdown phenomena in liquids [39–43]. However so far, a comprehensive theory of the breakdown in liquid does not exist yet and sometimes one possible mechanism proposed in one experiment is inconsistent with observations in another set of devices. Since very different experimental setups are used individually, it is very hard to compare and conclude from those results. In this article, we aim to highlight and analyse the current status of possible streamer discharge breakdown mechanisms in liquids that are available in published literatures, in particular, underlying physics and our understanding on these mechanisms are represented.

The paper is organised as follows. In Section 2, the basic physics of streamer and its classification are briefly introduced. Then, typically observed streamer photographs in various dielectric liquids are present, in particular, different characteristics with

respect to the polarity of electrodes is summarised. We present available theories of streamer discharge in liquids, in Section 3. Meanwhile, each mechanism suited for certain types of discharges is discussed in detail. Finally, conclusions and future outlooks are drawn in Section 4.

2 Streamer and experimental observations in liquids

2.1 Streamer and its classification

When an electron is inserted into an external electric field, it is accelerated by the electric force acting on it. If the background field exceeds the breakdown threshold of the medium, the electron with a certain kinetic energy can ionise neutral molecules by impact ionisation, in gases, this kinetic energy is typically above 12 eV. As long as the space charge produced by the impact ionisation does not influence the background electric field significantly, we call it as 'avalanche', as shown in Fig. 1. As the ionisation density grows, the electric field at the tip of avalanche is enhanced eventually, but it is suppressed in the ionised interior. In this manner, long ionised channels, so-called 'streamers', can initiate. According to the Raether–Meek criterion [1], the transformation from an avalanche to streamer occurs when the total number of free electrons reaches 10^8 – 10^9 , or

$$\alpha_{\text{eff}} d \simeq 18 - 20, \quad (1)$$

where, α_{eff} is the effective ionisation coefficient and d is the length. The time of the avalanche to streamer transformation was estimated recently in standard temperature and pressure air [44], according to

$$t_{\text{RM}} \simeq 18/\alpha_{\text{eff}} v_d, \quad (2)$$

where v_d is the drift velocity of electrons. Effects on the streamer formation of photoionisation, background ionisation and attachment/detachment process between free electrons and oxygen molecules were discussed as well [44].

According to the streamer propagation direction, positive (cathode-directed) and negative (anode-directed) streamers are classified. Streamer is filled with conducting plasma in the interior and certain amount of free electrons have to exist in the high field

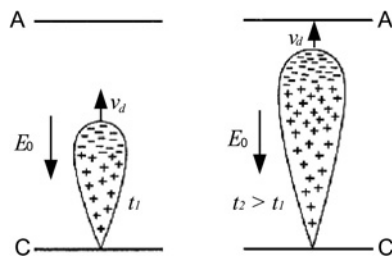


Fig. 1 Schematic of an electron avalanche at two consecutive moments of time. E_0 is the background electric field and v_d is the velocity of motion of the avalanche head. The figure is reproduced from [1]

region (close to the streamer head) to sustain its extension. Negative streamer propagates in the direction of the electron drift, so, electrons in the channel can directly supply to the streamer head. However, positive streamer propagates in the direction of the ambient electric field (against the electron drift direction), therefore, a source of electrons ahead of the streamer is required, see Fig. 2. Understanding where those source electrons come from is more critical for positive streamers than negative ones, since the former ones need them both for ignitions and for propagations.

2.2 Streamer observations in liquids and polarity effects

In this section, we show some typical structures of positive and negative streamers in liquids and the differences regarding the impact of the electrode polarity are present.

As shown in Fig. 3, negative and positive streamers ignite from a needle electrode selected from previous experiments are present, in different liquids: liquid nitrogen (Figs. 3a and b); transformer oil (Figs. 3c and d) and water (Figs. 3e and f). Surprisingly, streamer discharges in different liquids present very similar phenomena, although the geometry, composition of liquid molecules and pressure are entirely different. However, differences in patterns and structures of streamers from different polarised electrodes are seen significantly. Streamer has a thicker 'root' (or 'bushy' or 'mushroom-like') structure around the negative needle electrode, while many filamentary discharge channels present if the needle electrode is positively polarised. Actually, there are many interesting polarity effects observed and detected in previous experiments, mainly due to the differences in mobilities between electrons and ions (ion moves very slow due to its large mass) [49]. We here briefly summarise the observed polarity effects as follows:

- (i) *Size*: The diameter of the negative streamer channels is wider than positive streamers. As we see in Fig. 3, streamers from a negative needle electrode have a thicker root and bigger radial size, while streamers from a positive needle electrode are thinner and more filamentary.
- (ii) *Velocity and mode*: Streamers igniting from a positive needle electrode form earlier in time, move faster and do not tend to exhibit velocity saturation with the electric field amplitude [50]. However, the velocity of negative streamers was found to be not

affected by pressure and is orders of magnitude lower than positive ones. In addition, streamers originating on a positive needle electrode usually present two or more propagation modes [41, 51, 52] with different velocities (e.g. 100 m/s, 1, 10 and 100 km/s associated with four modes in mineral oil [51]), while negative streamers typically have one mode and possibly the second with a velocity <1 km/s [52]. Analogous to gas discharge, the velocity differences between positive and negative streamers can be explained due to their different field enhancement ability, that free electrons in the interior of the negative streamer channel drift outwards and reduce the field focusing at the streamer tip, while positive streamers are relatively narrow and therefore enhance the electric field at the tips to higher values [53].

(iii) *Inception voltage*: Streamers originating on a positive needle electrode require lower voltage and are frequently observed [36, 49]. This is a bit difficult to be understood, one might expect negative streamers from the needle can be easier due to possible field emission of free electrons from the cathode. However, as we explained above, the diffuse manner of the negative streamer weakens its electric field enhancement at the tip, thus, larger electric field is needed to support its ignition and propagation. It is worth to mention that some experiments in liquids showed opposite observations, for example, Fig. 2 in [54] and references therein. Remark that those experiments are performed under different conditions and at different groups, in addition, the inception voltage is also related to the stochastic event of having a starting electron available and suitable positions that are close to the pin electrode. Therefore, we would keep this question open and further detailed investigations are requested.

(iv) *Luminescence*: Almost in all experiments, positive streamers were observed to be brighter than negative streamers. One reason might be the effect of electron-ion recombination process. As present in Section 2.1, electrons spread out from the negative streamer tips but flow back to the channel for positive streamers. Since many ions exist in the streamer channel due to their low mobility, significant electron-ion recombination events occur in the positive streamer channels, leads to luminosity on their paths.

3 Streamer discharge mechanisms in liquids and discussions

Investigations on streamer discharge mechanisms in liquids started almost the same time as in gases and in solids. However, understanding streamer physics in liquids is more complicated, because liquids are denser than gases, softer than solids but lack of regular internal lattice structure. Additionally, properties (e.g. contamination, purity, conductivity) of liquids are easily affected by the environment, which makes the issue even difficult. Generally, scientists propose one or more mechanisms that can explain observations from their discharge systems, thus, many theories have been reported previously. However, as we mentioned early, since their experimental setups are very different, in most time, we cannot apply the mechanism from one experiment to another directly. In this section, we collect some possible

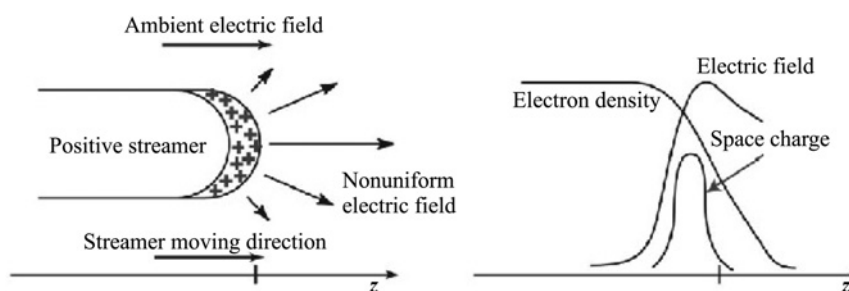


Fig. 2 Diagram of a positive streamer propagation in an ambient field. The figure is reproduced from [45]

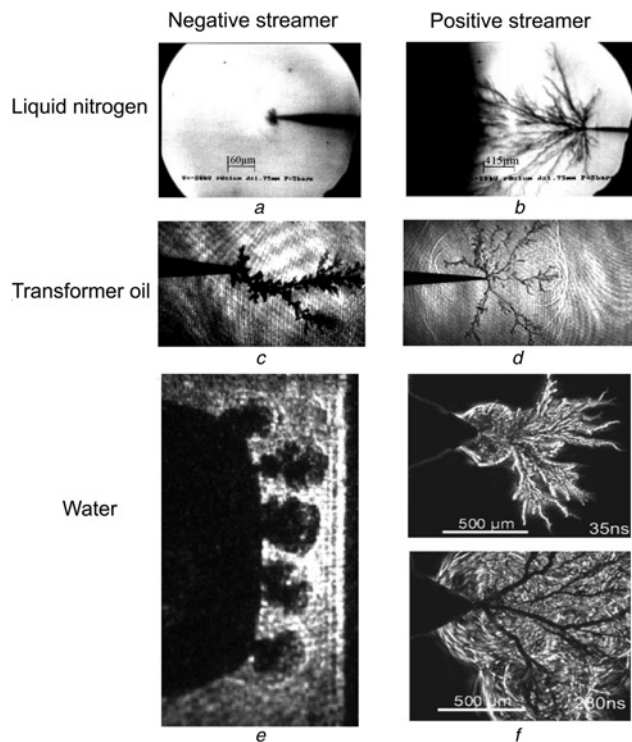


Fig. 3 Shadowgraphy of negative (left column) and positive (right column) streamers in different liquids: (a) and (b) in liquid nitrogen, (c) and (d) in transformer oil and (e) and (f) in water

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]

mechanisms from literatures that are relatively well accepted. We remark that we focus on analysing the applicability of those theories, but keep in mind that not all possible mechanisms are present here.

3.1 Direct impact ionisation

This mechanism assumes streamer formation in liquids is analogous to gaseous media. It is probably the most difficult one, because the liquid density is typically 10^3 – 10^4 orders of magnitude higher than in ambient pressure gases. Extensive scattering and very short mean free paths of electrons reduce strongly the possibility of reaching ionisation energy threshold. Additionally, in polar liquids (e.g. in water), electrons are to be solvated by the liquid molecules very rapidly within picoseconds time scales [34, 55], which makes free electrons even harder to sustain.

Direct impact ionisation mechanism may occur if some strict conditions are fulfilled. The first possibility is applying fast and extensive electric fields on the electrode, so free electrons are able to be accelerated to the ionisation energy threshold in a short time and length. As present in [39], a pulsed power system is used and the maximum electric field can reach ~ 220 MV/cm (670 Td) with an ultra-short rise time of a few picoseconds. The second possibility is creating a low density region, so, the impact ionisation can occur in the region with a longer mean free path

and lower ionisation threshold. This low density region can arise from pre-existence of gas bubbles, localised internal vaporisation, molecular decomposition or mechanical movement and extra [34, 35]. Some possible mechanisms for generating low density regions in liquids are also discussed in the following sections.

3.2 Gas bubble theory

As introduced in [56], the phase of liquids can become unstable and gas channels are able to form along the electric field lines. Since the density in the interior of the gas channel is lower, impact ionisation and avalanche are easier to happen than in dense liquid, if some free electrons exist in particular close to high electric field regions. At some point, micro discharges can form in the gas channels and gain more energy. So, discharges in micro gas bubbles are regarded as charge sources for the breakdown in liquids. Many experimental observations are consistent with these mechanisms that streamer are ignited from a gaseous phase, but it is not completely sure how gas bubbles are present. In general, the ‘bubble’ theory assumes (a) pre-existence of micro bubbles and/or (b) formation of bubbles. Gas bubbles usually exist in non-degassed liquids or in liquids with dissolved gas, note that small (nanoscale) gaseous pockets may always present even though the liquids were carefully degassed. The formation of bubbles can be a result of many possible reasons, for example, local joule heating in high electric field region that is close to the needle electrode, electrostatic expansion of pre-existing micro-bubbles and electrochemical effect [34, 35]. As an example, in Fig. 4, we present a recent schematic illustration of positive streamer initiation and propagation process underwater [41], which takes advantage of the gas bubble theory. Gas bubbles are first generated close to the needle electrode, due to the effect of joule heating. Weak discharges occur in the gas bubbles and lead to the formation of protrusions acting as a virtual sharp electrode. Then, streamer discharge in the water can carry on, since the electric field at the tip of the protrusions can exceed 10 MV/cm that is required for the ionisation event in water.

On the other hand, discharges in water without formation of gas bubbles were observed as well in experiments. As shown in [39], that a pulsed power system with ultra-short rising time and large amplitude electric field is used (33–220 kV pulse amplitude, 0.5–12 ns pulse duration and 150 ps rise time). They concluded that discharges are able to form on picoseconds time scale and gas bubbles are not need, or in strict speaking, no gas bubbles with a significant size are detected prior to their discharges.

So, gas bubble theory has generally been accepted for discharges with large pulsed widths (microseconds or longer) in liquids that allows the formation of gas bubbles. For the pulsed discharges on picoseconds or sub-nanoseconds time scale, the gas bubble theory might have to be re-evaluated.

3.3 Electric field dependent molecular ionisation

Electric field dependent molecular ionisation, also called field ionisation [57, 58], is a purely electric field driven process. Therefore, no free electrons are needed initially, which is quite different from direct impact ionisation mechanism. As illustrated in Fig. 5, when an extremely high electric field is present, an electron can be extracted from a molecule, resulting in both a free electron and an ion. Since the mobility values of free electrons and ions are very different, this field ionisation process will lead to significant net charge separation, in particular, within the timescales of streamer initiation and propagation.

Note that this mechanism has been widely used in models of streamer discharges in different dielectric media, for example, in transfer oil [18, 60, 61], in water [49] and in supercritical nitrogen [62]. The major difficulty of models with field ionisation process is the lack of theory in liquid state. Currently, most models use Zener theory to describe the field ionisation process in liquid, note that Zener theory has been developed previously for simulating electron tunnelling effect in solid semiconductors with a crystal

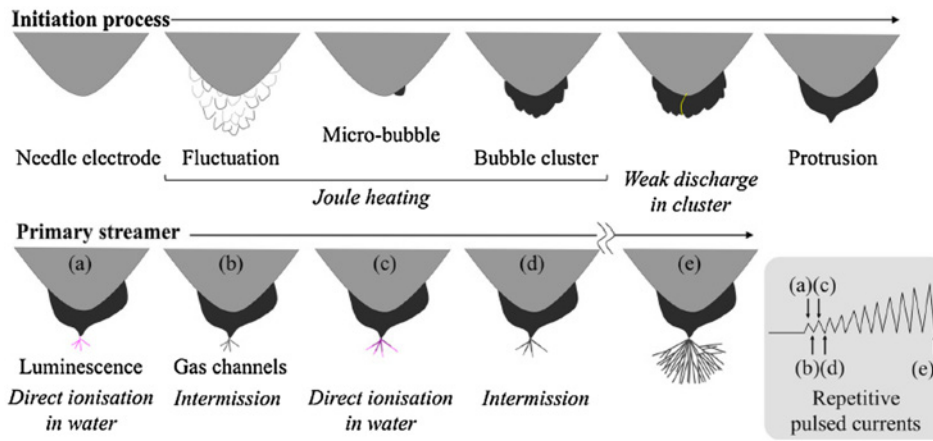


Fig. 4 Schematic view of positive streamer initiation and propagation process in water, which takes gas bubble theory. The figure is reproduced from [41]

structure [63]. According to Zener theory, the field ionisation rate G_I is determined by the following theory

$$G_I(|E|) = q^2 n_0 a |E| h^{-1} \exp(-\pi^2 m^* a \Delta^2 / q h^2 |E|), \quad (3)$$

where q is the electronic charge, h is the Planck's constant, $|E|$ is the electric field, n_0 is the number density of the molecules that field ionisation can occur, a is the molecular separation distance, Δ is the molecular ionisation energy and m^* is the effective electron mass in liquid. Equation (3) shows that the field ionisation rate significantly depends on various parameters, for example, the electric field stress, molecule density (pressure), ionisation potential and so on. To give a quantitative impression, we present a comparison of electron generation rate as a function of liquid pressures in Fig. 6, direct impact ionisation and field ionisation are both calculated. The medium we consider is supercritical nitrogen with 50 ppm impurities that field ionisation can easily occur. Other parameters can be found in [62]. It clearly shows that when the electric field is lower (5 MV/cm), the direct impact ionisation always dominates and the effect of field ionisation is very weak. Furthermore, if the electric field reaches 10 MV/cm, electron generate rate due to direct impact ionisation is still larger than field ionisation when $p < 50$ bar. However, when $p > 50$ bar, field

ionisation starts to produce the majority of free electrons and the effect of direct impact ionisation becomes small.

In conclusion, field ionisation mechanism has many advantages to explain the streamer initiation in liquids, since free charges are not required initially and impurities having lower ionisation potential might always exist no matter how much effort is spent for purification. However, be aware that field ionisation only dominates in high electric field region, which more likely exist close to the needle electrode or at the tip of streamers. The main drawback of this mechanism is lack of comprehensive theoretical data in liquids and experimental diagnostics on it is also very difficult to perform.

3.4 Electric field dependent ionic dissociation

Electric field dependent ionic dissociation is very similar to the field ionisation process, in principle. As shown in Fig. 7, this theory assumes liquid contains a certain concentration of neutral ion-pairs and free positive and negative ions. If the electric field is lower, the free charge concentration is low and the majority in the liquid is neutral ion-pairs. On the other hand, if an extensive electric field is used, the concentration of free charges increase due to the effect of electric field driven dissociation of the neutral ion-pairs.

As a result of the field driven dissociation process, the conductivity of the liquid increases significantly, thus, is in favour of the breakdown process. However, due to the low mobility of the heavy ions, the charge separation effect within the timescales of streamer formation is very weak, so, we do not expect this mechanism helps much at the streamer initiation stage.

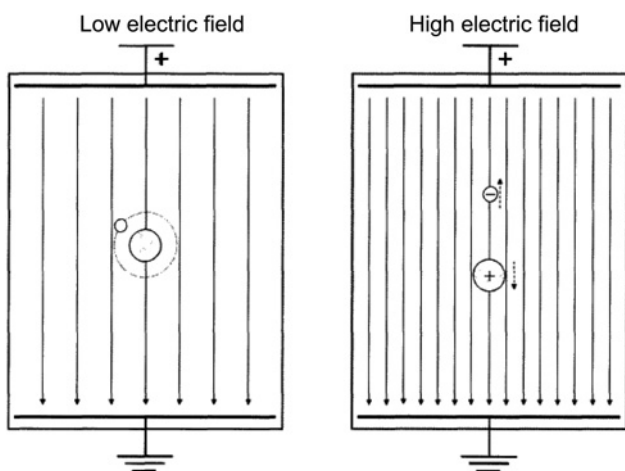


Fig. 5 Illustration of electric field dependent molecular ionisation process. Left: No molecular ionisation takes place at low electric field levels. Right: Generation of a free electron and positive ion from a neutral molecule in the liquid bulk, at high electric field levels. The figure is reproduced from [59]

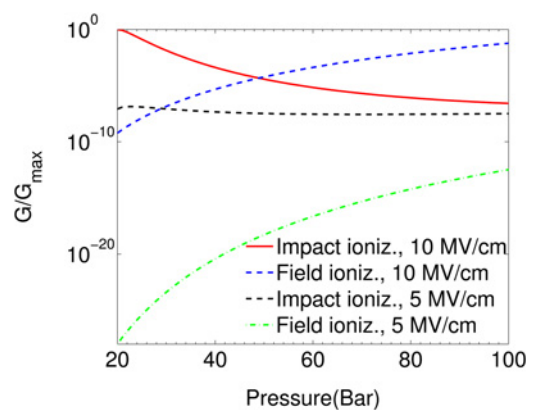


Fig. 6 Comparison of the electron generation rate as a function of pressure in liquid nitrogen. We assume 50 ppm impurities exist in the liquid that field ionisation can occur

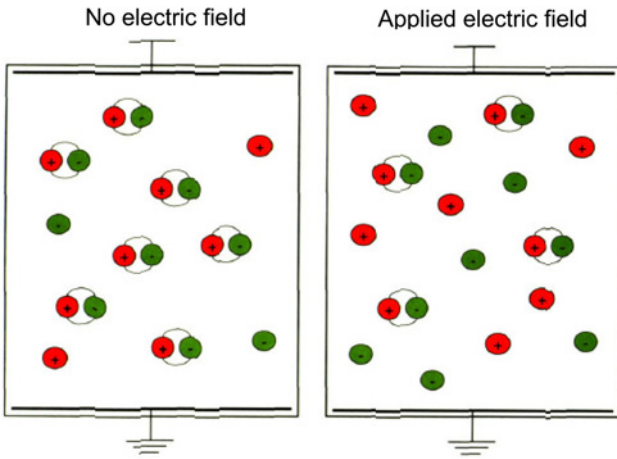


Fig. 7 Illustration of electric field dependent ionic dissociation process. Left: the majority in liquid is neutral ion-pairs when the electric field is low. Right: the concentration of free charge and conductivity of the liquid increase if an extensive electric field is used. The figure is reproduced from [59]

3.5 Auger process

Auger mechanism was previously discussed in [34, 35, 64] and energetic electrons are required initially. Those electrons would be trapped quickly (especially in polar liquids) by molecules. Meanwhile, excess energy from the incoming charge is absorbed by electron lying in excited states of the field-stressed molecule. Finally, two or even more electrons would be released from the capture site. So, a mass of free charged are created in this manner and charge separation can occur, inducing electric field enhancement and increasing conductivity of the liquid. In this manner, Auger process can continue and provide free charges continuously.

It is worth to mention that where initial energetic electrons come from is a puzzle. Usually, those electrons are regarded as a result of field emission and/or tunnelling effect from the cathode. So, this mechanism is more suited for explaining streamers ignited from a negatively polarised needle electrode.

3.6 Electrostriction mechanisms

Recently, a new mechanism was proposed for explaining sub-nanosecond pulsed breakdown in liquid dielectrics, so-called ‘electrostriction’ mechanism [40, 65–70]. The schematic view of this mechanism is shown in Fig. 8. In a needle-to-plane discharge system, non-uniform high electric field exists in the region close to the needle. Dielectric stress is affected by such large electric field gradient induced ponderomotive (electrostrictive) forces, leads to deformation and discontinuity of the liquid towards the region of high electric field. As a result, nano-sized pores or voids are

formed in the region of high electric field. In particular, those voids are elongated along the electric field lines, instead of spherical, thus, provide sufficiently long path for electrons to gain enough energy from the background electric field and to trigger impact ionisation process. Note that ‘voids’ here are different from the gas bubbles in Section 3.2, while ‘voids’ represents liquid-ruptures of smaller size and lower density. The electrostrictive force acting on the liquid is according to

$$F \simeq \frac{\epsilon_0 (\epsilon - 1)(\epsilon + 2)}{2 \cdot 3} \nabla E^2, \quad (\text{in non-polar liquids})$$

$$F \simeq \frac{\alpha \epsilon \epsilon_0}{2} \nabla E^2, \quad (\text{in polar liquids}), \quad (4)$$

where ϵ_0 is the vacuum dielectric permittivity, ϵ is the relative dielectric constant of the medium, E is the electric field and α is an empirical constant for polar liquids.

From (4), we emphasise that electrostrictive mechanism is insensitive to the polarity of the electric field, and is particularly suited for sub-nanoseconds rising and strong non-homogeneous electric field in the vicinity of high-voltage needle electrode.

Note that if rate of the voltage rise is slow (microseconds), hydrodynamic pressure would counteract the electrostrictive effect and ‘voids’ may not form or take longer for critical size to be created. On the other hand, if the voltage rising time is ultra-short (picoseconds), the discharge may ignite from direct impact ionisation without phase transition or formation of nanoscale voids, as we introduced in Section 3.1.

3.7 Other mechanisms

In many aforementioned mechanisms, one difficulty is determining where the seed electrons are obtained before switching on the voltage signal. We remark that in gas discharge physics, photoionisation and background ionisation due to cosmic ray or radiation provide seed electrons for pre-breakdown. Similar mechanisms are also considered in liquids. It is true that there is no direct evidence of the existence of photoionisation during the breakdown in liquids, since it is very difficult to measure. However, light emission has always been observed and been linked to the propagation of streamers [71, 72]. Additionally, if streamers ignite on a negative electrode, field emission or tunnelling from the cathode might be an important source of electrons [59, 73].

4 Concluding remarks

Streamer-like discharges in liquids have been intensively investigated due to their interesting applications such as purification, decontamination, biomedicine, nanoscience and high-voltage insulator/switch. The emphasis of this review paper is on the streamer formation mechanisms directly in liquids.

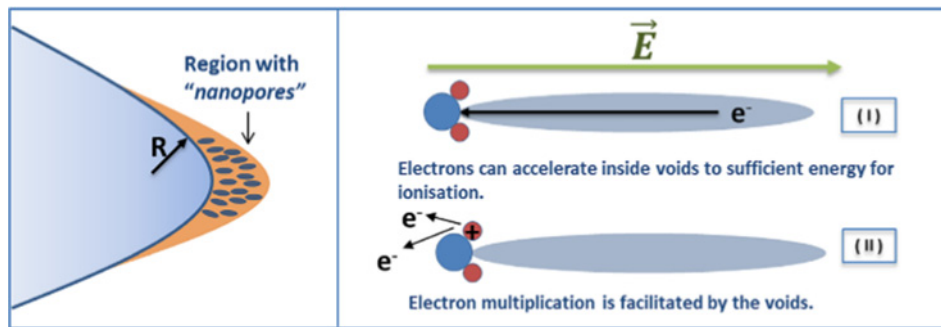


Fig. 8 Schematic of electrostriction mechanism for plasma initiation in water by nanosecond pulses in needle to plane electrode geometry. The figure is reproduced from [66]

The basis physics of streamer theory and experimental observations of streamers in various liquids related to polarity effect are present. We then discuss in detail available theories of streamer formation from the literatures, including direct impact ionisation, gas bubble, electric field dependent molecular ionisation/ionic dissociation, Auger process and electrostriction mechanisms. In particular, we specify the applicability and limitation of the each theory, according to our understanding.

So far, a comprehensive theory of streamer discharge in liquid does not exist and there are many questions need to be answered. The situation becomes even worse, since many aspects affect the understanding of discharges in liquids, for example, the purity, hydrostatic pressure, viscosity and conductivity of the liquid, material and polarity of the electrode. As an outlook, further experimental and numerical explorations are extensively required. Major difficulties of experiments on discharges in liquids include expensive cost and discharges are fast and small, thus, reliable data is not easy to measure. Plasma discharge modelling probably can play increasing roles in exploring the underlying physics in liquids, since it allows for neglecting other mechanisms and focusing on one theory of interest, but the lack of reliable cross-sections and parameters reduces the accuracy of the modelling. On the other hand, existences of aforementioned puzzles and difficulties make the topic of streamer discharge in dielectric liquids more interesting and attract increasing scientific attentions to this field.

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