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FAST TRACK COMMUNICATION

Structure of positive streamers inside gaseous bubbles immersed in liquids

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Abstract

Electric discharges and streamers in liquids typically proceed through vapour phase channels produced by the streamer or in gaseous bubbles. The bubbles can originate by enthalpy changes produced by the discharge or can be artificially injected into the liquid. Experiments on streamers in bubbles immersed in liquids have shown that the discharge propagates either along the surface of the bubble or through the volume of the bubble as in conventional streamer propagation in air. In this paper we report on results of a computational investigation of streamer propagation through bubbles immersed in liquids. We found that the dielectric constant of the liquid in large part determines the path the streamer takes. Streamers in bubbles immersed in a liquid with a high permittivity preferentially propagate along the surface of the bubble. Liquids with low permittivity can result in the streamer propagating along the axis of the bubble. The permittivity at which this transition occurs is a function of the applied voltage, size of the bubble and the conductivity of the liquid.

1. Introduction

Electric discharges in liquids, and streamers in particular, are being intensively investigated due to their wide-ranging applications, from removal of volatile organic compounds (VOCs) from water to surgical instruments [1–5]. In most cases, streamers do not propagate directly through the liquid phase. Rather breakdown likely occurs inside bubbles and near gas–liquid interfaces. Enthalpy changes produced by the discharge result in further vaporization of the liquid, creating gaseous channels. Radicals produced by the plasma in bubbles interact with chemical components in the liquid by transport through the gas–liquid interface. As such, more efficient remediation of, for example, VOCs in liquids may be obtained by injection of bubbles in the vicinity of point-electrodes. This helps facilitate the formation of in-bubble plasmas in the electric field enhanced region by decreasing the applied voltage required for breakdown [3].

The paths streamers take in bubbles, particularly near point-electrodes, are not clearly defined. Bruggeman showed that streamers preferably take paths along the surface of the bubble when the bubble is immersed in a high dielectric constant liquid such as water [4, 5]. Gershman *et al* noted that discharges in bubbles are analogous to dielectric-barrier discharges where the water serves as a dielectric for reducing current [3]. The discharge deposits charge on the surface of the bubble which can affect its path and which can also produce a reverse-polarity discharge. More details of the theory and experimental results on streamer inception and propagation in liquids can be found in [6].

In this paper, we discuss results from a modelling study of the propagation of positive streamers in bubbles immersed in a liquid. The bubbles contain atmospheric pressure humid air. We found that the dielectric constant and conductivity of the liquid largely determine the path and pattern of streamer propagation. Streamers in bubbles immersed in liquids with a high permittivity preferentially propagate along the surface of the bubble. Liquids having a low permittivity produce streamers that propagate along the axis of the bubble. The higher the conductivity of the liquid, the smaller the permittivity at which this transition occurs. The particular

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pattern of the streamer path is also a function of the bubble size, applied voltage and its polarity, as well as the mean free path of ionizing photons. The behaviour of streamers in bubbles is similar to that of dielectric-barrier discharges where the liquid serves as a lossy dielectric electrode.

2. Description of the model

The model, *nonPDPSIM,* and the reaction mechanism used in this study are the same as those described in [7] and so will be only briefly described here. *nonPDPSIM* is a two-dimensional simulation in which Poisson's equation for the electric potential and transport equations for charged and neutral species are solved. The electron temperature, T_e, is obtained by solving an electron energy conservation equation with transport and rate coefficients coming from local solutions of Boltzmann's equation. Radiation transport and photoionization are included by implementing a Green's function propagator. The model geometry is shown in figure 1(*a*) and is symmetric across the centre-line. The numerical grid uses an unstructured mesh with triangular elements and refinement regions to resolve the details of the electrode tip and the bubble. The mesh consists of approximately 6000 nodes, of which about 4000 are in the plasma region inside the bubble. A positive corona discharge is sustained between a rod encased in a dielectric having an exposed edge with a radius of curvature of 0.07 cm and a ground plane 2–4 mm away. The tip of the electrode is immersed in a liquid whose dielectric constant and conductivity were varied. The plasma transport equations are solved only inside the bubble. Inside the liquid, a conductivity, σ, is specified and charge densities are solved for by using the equation $\partial\rho/\partial t = -\nabla \cdot \vec{j} = -\nabla \cdot \sigma \vec{E}$. Charge is allowed to accumulate at the bubble–surface interface consistent with the incident fluxes from the plasma and the currents through the liquid.

The gas mixture is $N_2/O_2/H_2O = 55/15/30$ with an ambient pressure of 1 atm. The radii of the bubbles were 450 to 900 μm. For the cases discussed here the mean free path of ionizing photons was 100 μm. Seeding the streamer with initial electrons likely includes contributions of electron injection into the bubble from the liquid. However, in this study, to initiate the discharge, a small cloud of seed electrons with a radius of 500 μm and a peak density of 10^8 cm^{-3} was placed at the upper boundary of the bubble near the electrode. This assumption does not change the end result of streamer evolution, but does simplify the problem. We investigated cases where the bubble is totally immersed in the liquid with a thin layer of liquid between the bubble and electrode; and cases where the tip of the electrode is inside the bubble. In these simulations, we intentionally eliminated processes which may take place on the liquid–gas boundary (e.g. secondary electron emission and evaporation) in order to isolate the effects of changing the permittivity and conductivity of the liquid.

Our study addresses streamer evolution on the nanosecond time scale during which the bubble can be considered a static structure. We acknowledge that bubbles can evolve under the influence of electric fields and discharge heating (e.g. elongation due to the applied electric field and evaporation with subsequent expansion); however, these effects take place on a microsecond time scale and so are not addressed here. For a similar reason, the effect of electric field-dependent permittivities of the liquid were not taken into consideration. Although the conductivity of the liquid was included, the liquid was assumed to be non-ionizing media.

3. Streamers inside bubbles

It is well known that the electric field inside a spherical cavity (e.g. a bubble) immersed in a dielectric medium having a uniform electric field is also uniform and of magnitude $(\varepsilon/\varepsilon_0 + 2)E/3$, where $\varepsilon/\varepsilon_0$ is the relative permittivity of the dielectric and E is the uniform field in the dielectric [8]. The distribution of electric potential and field for a bubble in a divergent electric field differs significantly from that for a uniform external field. For example, the electric potential and field near the electrode tip adjacent to a 700 μm bubble are shown in figure 1(*b*). Refraction of the electric field lines at the bubble–liquid interface produces electric field enhancement which increases with increasing $\varepsilon/\varepsilon_0$ of the surrounding liquid while excluding potential from the bubble. These trends in large part explain the behaviour of streamers in bubbles immersed in liquids.

Electron density, T_e and positive space charge during positive streamer propagation inside a bubble are shown in figure 2 for the two limiting cases of liquids having $\varepsilon/\varepsilon_0 = 2$ (e.g. a fluorocarbon liquid) and 80 (e.g. water). The conductivity of the liquid is $\sigma = 10^{-7}\ \Omega^{-1}$ cm^{-1}. In the case of the low dielectric constant liquid, the streamer develops as a single on-axis filament with an avalanche front of increasing speed as would be the case in open air [9]. In traversing the bubble, the electron density in the avalanche front increases from 6×10^{14} cm^{-3} to 5×10^{15} cm^{-3} and T_e increases from 9.8 to 13 eV. The positive space charge outlines the avalanche front [10]. The avalanche front collides with the cathode side of the bubble, depositing its charge on the liquid surface similar to a dielectric barrier discharge.

Streamer propagation in the bubble immersed in the liquid having $\varepsilon/\varepsilon_0 = 80$ (essentially de-ionized water) is markedly different. Refraction produced by the curved dielectric boundary directs electric field lines towards the surface of the bubble. The streamer is initiated in the geometrically enhanced electric field at the tip of the electrode and first attempts to propagate along the axis. The concentration of electric field lines along the boundary diverts the streamer to propagate in a thin layer along the surface. The positive space charge shows that the axial streamer stalls out as the boundary-hugging avalanche takes over. As the boundary-hugging avalanche produces a plasma column along the surface of the bubble, charge is deposited on the boundary. There is additional electric field enhancement at the head of the avalanche as the conductive plasma channel forms, which produces T_e of 24–47 eV, with a peak electron density of $(3–8)\times10^{16}$ cm^{-3}. These characteristics are similar to those found in conventional surface discharges [11]. The streamer circumnavigates the

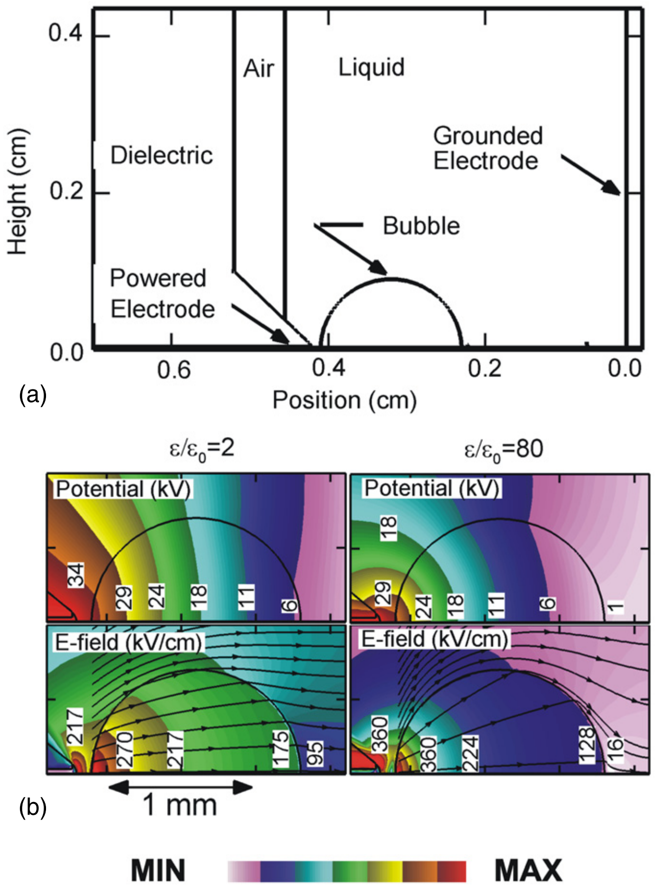

Figure 1. Schematic of the geometry and electrical properties. (*a*) The computational domain in the vicinity of the bubble, symmetric across the lower boundary. (*b*) Electric potential and electric field streamlines for a bubble 700 μm in radius immersed in a liquid with a (left) low dielectric constant ($\varepsilon/\varepsilon_0 = 2$) and (right) high dielectric constant ($\varepsilon/\varepsilon_0 = 80$). The tip of the electrode is at the left of the frames. Refraction of the electric field lines on the bubble–liquid interface results in more divergent fields for higher $\varepsilon/\varepsilon_0$. The streamlines show the direction of the electric field but not field magnitude.

bubble in a shorter time than required by the axial streamer to cross the gap, a consequence of the higher electric field along the boundary.

The electron density, electron impact ionization source (S_e) and positive space charge are shown in figure 3 for $\varepsilon/\varepsilon_0 = 2$ to 80. Typically, two streamers simultaneously develop inside the bubble, one propagating along the axis and one along the surface. Which streamer ultimately dominates is a function of $\varepsilon/\varepsilon_0$ of the liquid. For lower permittivity ($\varepsilon/\varepsilon_0 < 8$) the axial streamer develops faster than its surface companion. With an increase in the dielectric constant ($\varepsilon/\varepsilon_0 > 8$), and a subsequent increase in the magnitude and degree of divergence of the electric field, the surface streamer intensity (electron density, impact sources and velocity) increases. Eventually, the boundary-hugging streamer dominates. In addition to the geometrical refraction which reduces the on-axis electric field, the boundary-hugging streamer gradually screens the electric field inside the cavity helping to suppress the development of the axial streamer. For higher dielectric constants the two streamers (axial and surface) develop nearly independently, with the axial streamer stalling prior to crossing the bubble. The peak electron density monotonically increases with increasing $\varepsilon/\varepsilon_0$ of the liquid.

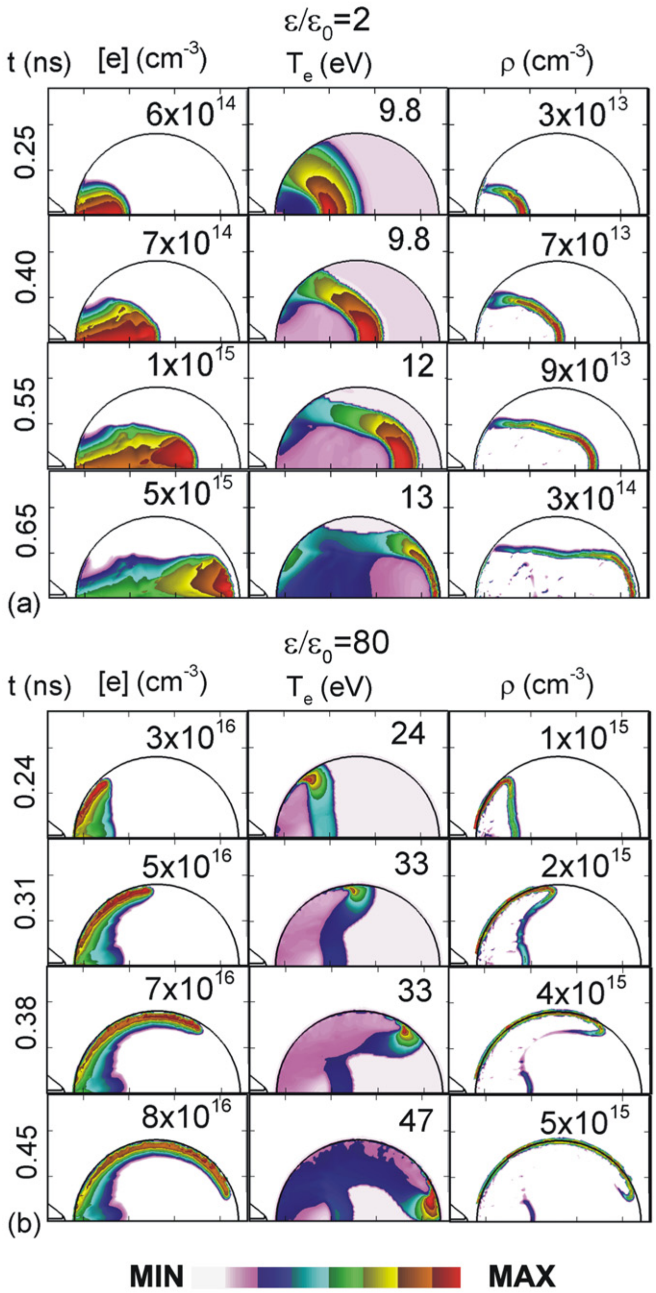

Figure 2. Streamer properties inside a bubble 900 μm in radius immersed in a dielectric liquid with (*a*) $\varepsilon/\varepsilon_0 = 2$ and (*b*) with $\varepsilon/\varepsilon_0 = 80$. The conductivity $\sigma = 10^{-7}\ \Omega^{-1}\ \mathrm{cm}^{-1}$. The electron density (3 decade log plot), positive space charge (2 decade log plot) and electron temperature are shown for different times. The maximum values are noted in each frame. In the low $\varepsilon/\varepsilon_0$ liquid, the streamer develops as a single filament propagating along the axis with properties similar to conventional streamers in unconfined gases. In the high $\varepsilon/\varepsilon_0$ liquid, the streamer propagates along the bubble–surface depositing charge on the boundary.

In addition to the $\varepsilon/\varepsilon_0$ of the liquid, the structure of streamers inside bubbles also depends on the voltage, conductivity of the liquid, size of the bubble and its position

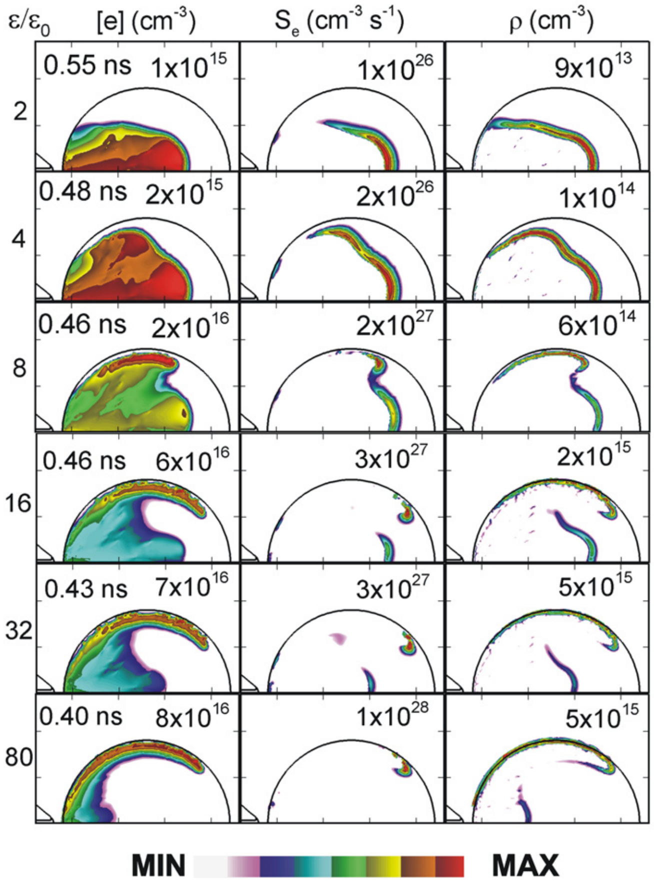

Figure 3. Streamer properties inside a bubble 900 μm in radius immersed in liquids with different $\varepsilon/\varepsilon_0$. Electron density (3 decade log plot), electron impact source (4 decade log plot) and positive space charge (2 decade log plot) are shown for the same time. Typically, two streamers develop inside the bubble—along the axis and along the surface. The final pattern depends on $\varepsilon/\varepsilon_0$, with higher values favouring the surface discharge.

with respect to the tip of the electrode. For example, streamer properties are shown in figure 4(*a*) for a liquid having $\varepsilon/\varepsilon_0 = 2$ and 80 for $\sigma = 10^{-2}\,\Omega^{-1}\,\mathrm{cm}^{-1}$. When immersed in a conductive liquid, the surface-hugging streamer develops at a lower value of $\varepsilon/\varepsilon_0$. Due to the high conductivity of the liquid, negative charges from the liquid accumulate on the outer boundary of the bubble. As a consequence, electric field lines become more divergent as they approach the surface of the liquid. The pulse power format therefore becomes more important as the conductivity of the liquid increases as charges deposited by the streamer on the surface of the bubble will dissipate in time due to the finite conductivity of the liquid.

Streamer development does not dramatically change when the tip of the electrode sticks into the bubble as shown in figure 4(*b*). One difference is that the streamer is wider. Being closer to the electrode the streamer develops in a larger electric field which tends to broaden the streamer [12]. There is also little dependence of the streamer structure on the distance between the bubble and the lower electrode provided there is sufficient electric field to launch the streamer. This is shown in figure 4(*c*) where the lower electrode is an additional 2 mm away (total gap is 4 mm) and the applied potential is 80 kV

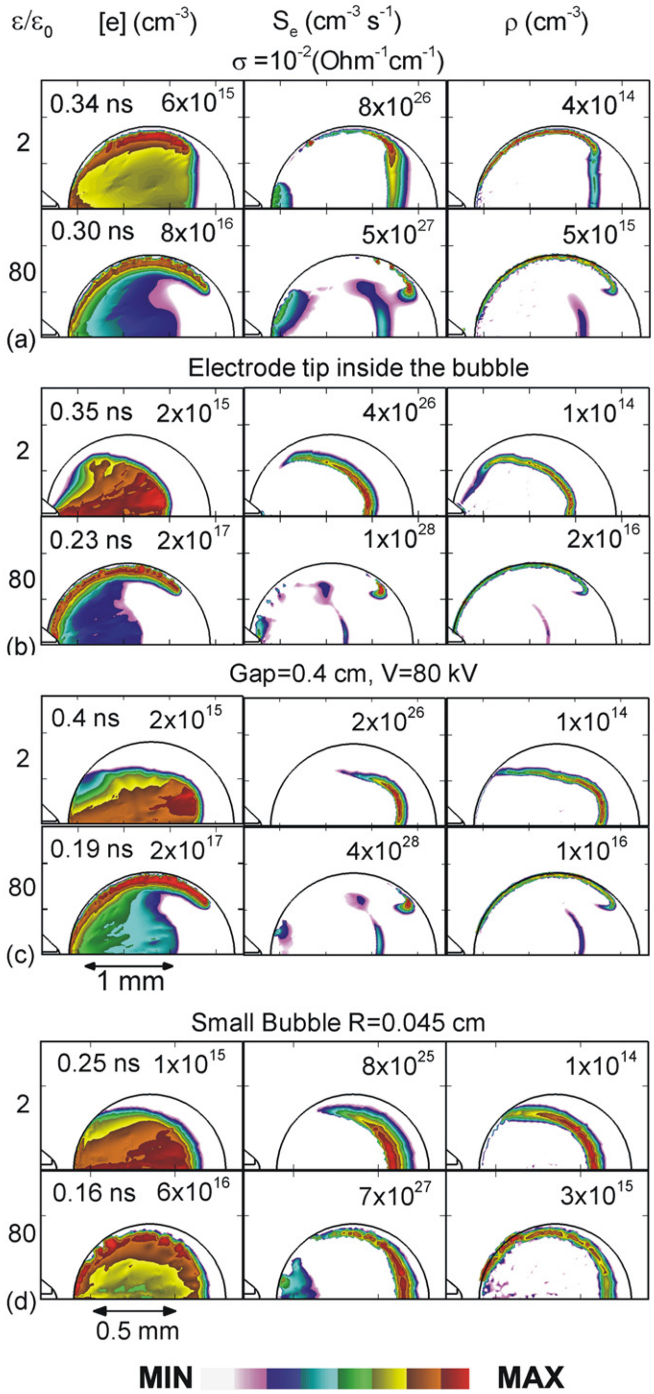

Figure 4. Electron density (3 decade log plot), electron impact source (4 decade log plot) and positive space charge (2 decade log plot) for (*a*) a liquid of high conductivity ($10^{-2}\,\Omega^{-1}\,\mathrm{cm}^{-1}$), (*b*) the electrode tip inside the bubble, (*c*) an electrode gap of 0.4 cm and (*d*) a smaller bubble of 450 μm radius. Streamers become boundary hugging at lower $\varepsilon/\varepsilon_0$ as the liquid conductivity increases. Streamer propagation is not particularly sensitive to the positioning of the electrode tip or location of the ground plane. When the natural streamer width becomes comparable to the size of the bubble, the surface hugging and axial streamers merge.

(to keep a constant average E/N across the gap). If the bubble is moved outside the divergent electric field at the tip of the electrode, the boundary-hugging streamer becomes less dominant.

The patterns of streamer development do depend on the size of the bubble, as shown in figure 4(d) for a bubble of 450 μm radius. In this case the bubble size is comparable to the natural width of the streamer. The axial and boundary-hugging streamers then merge, and there is less distinction between streamers sustained in bubbles immersed in liquids having low and high $\varepsilon/\varepsilon_0$.

4. Concluding remarks

We investigated the properties of positive streamers propagating inside bubbles immersed in liquids of varying dielectric constant and conductivity. We found that increasing $\varepsilon/\varepsilon_0$ and σ produces streamers that propagate along the surface of the bubble. This is in large part due to electric field enhancement that occurs on the boundary of the bubble at the curved interface across different dielectric constants. The surface-hugging streamer has many of the characteristics of a conventional surface discharge. The transition from an axial to a surface-hugging streamer additionally depends on the size of the bubble, the voltage, the mean free path of ionizing photons. As the voltage and mean free path for ionizing photons increases, the width of the streamer increases. So for a bubble of a given diameter, streamers are more likely to track along the surface. For the same reason (other conditions being equal) smaller bubbles and negative voltages (which produce wider streamers than positive voltages) are expected to produce surface-hugging streamers.

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