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The secondary ionization wave and characteristic map of surface discharge plasma in a wide time scale

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Abstract

The surface discharge is the elementary process in a broad range of low temperature plasma applications. Variation of voltage profiles with different time scales leads to the redistribution of deposited energy as well as electro-hydrodynamic forces of surface discharges, while the mechanism and scaling law is still unknown. On the basis of theoretical and numerical analysis, we show that a secondary surface ionization wave forms during the voltage rising slope when electron density decreases to a critical level while the voltage is still rising. A characteristic map of energy and electro-hydrodynamics force in time scales between 1 ns and 0.1 s at atmospheric pressure is proposed, opening the door towards the target-directed design of surface discharges.

1. Introduction

Surface discharges are widely observed and intensively studied by researchers in the community of low temperature plasma applications. A surface discharge can be generated between two electrodes separated by a dielectric barrier, by applying a high pulse or sinusoidal voltage on one of the electrodes, a surface ionization wave (SIW) with an ionization front of high electric field will appear and propagate along the dielectric surface, leaving a thin sheet region of plasma [2], this configuration is called surface dielectric barrier discharge (SDBD) [1].

The use of SIW has been the subject of many experimental and theoretical works since the 20th century due to its unique characteristics [3]. The SIW is a mixture of negative and positive ions and can be accelerated by the electric field, leading to the ionic wind above the surface for active flow control [4–6]. The fast heating energy released in SIW has been used for ignition in rapid compression machines [7, 8] and active icing control over airfoils [9–11]. The large discharge sheet make SIW an useful tool for surface treatment [12, 13]. The plasma region in the SIW body can be considered as a chemistry reactor producing specific species, e.g. in ozone generators [14] (widely used for room sterilization in current COVID-19 period), plasma activated water producers [15] and plasma jets [16, 17] (used for medical treatment of skins *et al*).

Despite the wide applications, the physics of SIWs, especially the knowledge of discharge dynamics, the thrust and deposited energy which are elementary for aforementioned researches, are not fully understood due to limited measurement techniques and theoretical basis in the past years. The lack of physical ground limits the targeted design of SIW devices in applications. The advances of fast imaging techniques, modeling theories and tunable power sources in recent years have allowed deeper insights into the discharge dynamics and hydrodynamics characteristics of the SIWs. The propagation of a SIW driven by a short nanosecond pulse is observed through optical spectrum emission and successfully reproduced by numerical simulations [18, 19]. A set of analytical solutions is built based on experiments and latest simulations [20–22].

In recent experiments [23, 24], an new voltage pulse generator with tunable rising slopes is used, secondary current spikes are observed during the voltage rising edge, which are different from the widely known secondary current spike in the voltage trailing edge when the electric field is reversed. This phenomena has not been observed before and the mechanism is not clear yet.

In this work we perform simulations to show the propagation of the SIWs in a recent experiment [23], i.e. in air at atmospheric pressure driven by voltages of different rising slopes. The model results give a good quantitative prediction and explanation of the formation of the secondary ionization wave during voltage rising slope. Based on the experimental data, simulations and the analytical solutions summarized in this work, we propose a general scheme of energy and electro-hydrodynamic force characteristics of surface discharge plasma as functions of voltage rising time and amplitude.

2. The formation of the secondary ionization wave

In the surface discharges driven by voltage pulses, the secondary breakdown has long been observed at the trailing edge of the voltage pulse: the electric field and current change the direction at this stage, leading to the generation of a secondary SIW of the opposite polarity near the electrode.

At specific conditions, there exists a secondary ionization wave having the same polarity of charged head and current with the first one. This phenomena often occurs when the voltage (hundreds of kV) rising time is hundreds of microseconds and the gap is long (~ 1 m) and was known as the stepwise leader. The mechanism is closely related to the gas heating and changes in the gas density [25, 26].

However the secondary SIW occurs in a much shorter time scale (hundreds of ns) even before the first wave ends. We notice that this time scale corresponds to the electron decay due to the dissociative recombination process [21]:

$$n_e(t) \approx \frac{n_{e0}}{1 + t/\tau_r} \quad (1)$$

where $\tau_r^{-1} = 1.4 \times 10^{-12} (300/T_e)^{0.5} n_e$ is the characteristic time of dissociative recombination of electrons with O_4^+ ions, n_{e0} is the electron density formed when the first ionization front passes, t is the time after the first ionization wave passed.

When the electron density in the surface plasma channel decreases to $10^{18} - 10^{19} \text{ m}^{-3}$, the in-channel region cannot shield the increasing external electric field. The high near electrode electric field plus the highly pre-ionized plasma trace left by the first ionization wave is possible to trigger a secondary ionization wave.

To formulate the critical electron density or time moment of the secondary SIW, the initial electron density n_{e0} can be expressed as [21]:

$$n_{e0} \approx \frac{\nu_{ic} V_h^2 \epsilon^2}{8\pi e \mu E_c^2 d^2} \quad (2)$$

where the ionization frequency ν_i can be closely approximated by the dependence $\nu_i = \nu_{ic}(E/E_c)^2$, $\nu_{ic} = 3.5 \times 10^{11} \text{ s}^{-1}$ independent from gas density, and $E_c = 277 \text{ kV cm}^{-1}$ in atmospheric pressure. $\epsilon = 4.3$ is the dielectric constant, $\mu \approx 0.06 \text{ m}^2 (\text{V s})^{-1}$ is the electron mobility. V_h is the potential of the first ionization head and can be expressed as $V_h = V_{bd} + kt_d$, where k is the voltage rising rate, $V_{bd} = 7.3 \times 10^4 (1 + 1/(\pi\epsilon)) h_d^{0.5}$ is the breakdown voltage, t_d is the discharge propagation time:

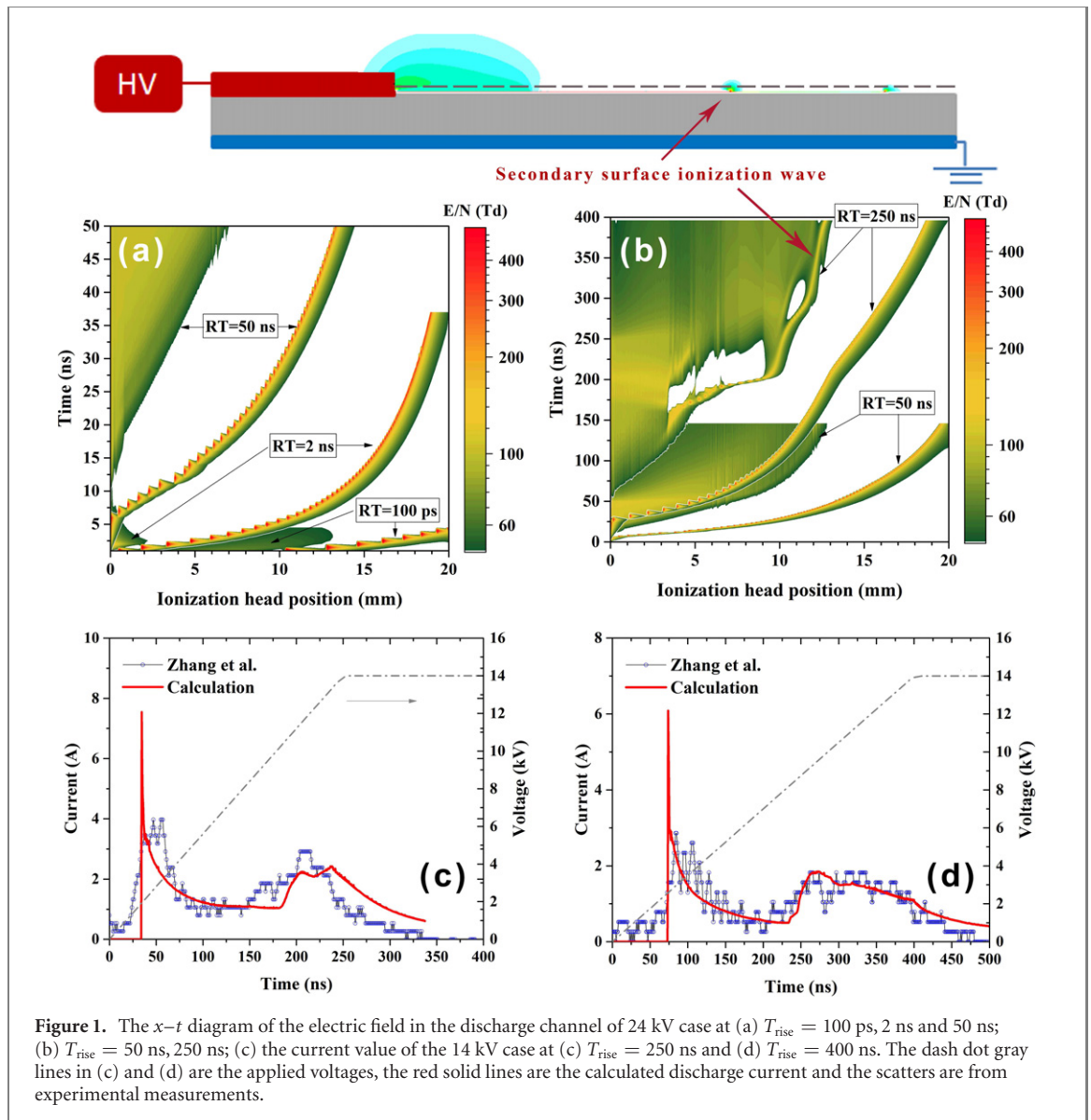
$$t_d \approx \frac{4h_d^2 \nu_{ic}}{\mu^2 E_c^2 \epsilon} \quad (3)$$

where h_d is the thickness of the plasma channel ($\sim 50 \text{ }\mu\text{m}$ in atmospheric pressure). At critical time moment $t = t_{cri}$, the electron density decreases to n_{ecri} when the secondary ionization wave occurs. Substituting equations (2) and (3) and $n_e(t) = n_{ecri}$ into (1) leads to a quadratic equation. The solution plus the time before breakdown gives the criteria time moment of the second SIW:

$$t_{cri} \approx 1.25 \times 10^{12} \frac{n_{e0} - 2n_{ecri}}{n_{ecri}^2} + \frac{V_{bd}}{k} \quad (4)$$

The condition of secondary SIW formation can then be expressed as $t_{rise} > t_{cri}$ or $n_e < n_{ecri}$ (on the basis of $V > V_{bd}$, the requirement for inception voltage has been implicitly embedded in equations (4) and (2)). It has to be noted that equation (1) is a rough approximation requiring the information of mean electron temperature T_e . The accuracy of the estimation of n_{e0} from equation (2) is affected strongly by the accuracy of V_h , n_{e0} *et al.* To have a more precise calculation for equation (4), detailed numerical simulations are necessary.

We now take the experimental parameters as the input of our discharge model and investigate the condition of the secondary SIW formation. The discharge is modeled with the classical drift-diffusion-reaction model for $N_2 : O_2$ mixture based on PASSKEY code [18]. Two voltage amplitudes are calculated: $V_{max} = 14 \text{ kV}$ as in the experiment [23] and $V_{max} = 24 \text{ kV}$.



The height of the simulation domain is 5 cm and its weight is 8 cm, sufficiently much larger than SIW scale that the background field can be decided by Neumann boundary conditions for the electric potential on the boundary. Together with the dielectric with permittivity of 4.3 and thickness of 1 mm, the electric field can be obtained by solving the Poisson's equation.

The equations are discretized on a static nonuniform grid. The grid is refined in the area where a streamer is expected to propagate. The size of the finest grid cells is $5 \mu\text{m}$. Away from the area of streamer propagation, grid cells exponentially increase in size up to 0.2 mm on the boundaries. To speed up the calculation, a smaller region with height of 0.1 cm and weight of 4 cm is used only to solve the drift-diffusion-reaction equations. Details of the numerical scheme, the chemical reactions, the coupling strategy with gas heating in this work and code validation have been presented in previous papers [18, 27] and will not be duplicated here.

The result of our simulations is shown in figure 1. Figures 1(a) and (b) show the x - t diagram of the field in the discharge channel ($25 \mu\text{m}$ above the dielectric) at 24 kV with different rising times ($t_{\text{rise}} = 100$ ps, 2 ns, 50 ns and 250 ns). For the same peak voltage, the ionization front propagates much faster with higher electric field at shorter rising times (the left panel). With the increase of the voltage rising time, the field near the electrode grows and a secondary ionization front appears starting from 150 ns in the $t_{\text{rise}} = 250$ ns case.

The calculated current values for the 14 kV case with $t_{\text{rise}} = 250$ ns and $t_{\text{rise}} = 500$ ns are plotted in figures 1(c) and (d) together with experimental measurements and applied voltage profiles. The starting moment and the amplitude of the secondary current peaks agree well with the analytical prediction and experimental measurements.

We take the electron density value from the discharge model as the input for equation (4) to calculate the criteria electron density n_{ecri} . At $V_{\text{max}} = 24$ kV and $t_{\text{rise}} = 250$ ns condition, $n_{e0} = 1.6 \times 10^{20} \text{ m}^{-3}$ and $t_{\text{cri}} = 150$ ns, this leads to $n_{\text{ecri}} = 5.1 \times 10^{19} \text{ m}^{-3}$. At $V_{\text{max}} = 14$ kV and $t_{\text{rise}} = 250$ ns condition we have $n_{\text{ecri}} = 4 \times 10^{19} \text{ m}^{-3}$. The same procedure applied to $V_{\text{max}} = 14$ kV and $t_{\text{rise}} = 400$ ns condition leads to $n_{\text{ecri}} = 3.7 \times 10^{19} \text{ m}^{-3}$. Thus, if the voltage rising time is longer than the time required for the electron density decaying to below $3 \sim 5 \times 10^{19} \text{ m}^{-3}$, it is possible that the secondary SIW appears.

As fast gas heating is an important process for nanosecond pulsed discharges, it is necessary to check the role of gas heating in formation of the second SIW. Fast gas heating affects the discharge by (i) decreasing neutral density thus increasing E/N ; (ii) changing reaction rates (especially quenching reactions of excited species that join associative ionizations). But both changes cannot sufficiently explain the occurrence of the secondary surface streamer. (i) The gas temperature increase almost immediately due to fast gas heating but it takes several microseconds and even mini seconds for the gas to expand, the change of neutral density is rather small in atmospheric pressure thus the change of E/N is also small in the studied time scale (100 ns). (ii) Although the change of reactions rates may affect the production of some excited species, as have been mentioned in the original manuscript, the plasma density is quite low thus the associative ionization is rather weak in atmospheric nSDBD. We have conducted an additional calculation by closing the gas heating module in the code and solve only for plasma equations (the Poisson's equation and the drift-diffusion-reaction equations). The secondary SIW still appears.

3. The energy deposition

Energy deposition is the pioneering process of gas heating/fluid responses and has been found to be related with the rising slope [28]. In general, shorter rising time leads to higher energy deposition (calculated from $j \cdot E$), but quantitative analysis is not available.

We calculate the total energy deposition for different rising slopes shown in figure 2. The upper panel shows the spatial distribution of deposited energy. The energy is distributed smoothly along the plasma channel at short pulses (figure 2(a) and [27]). At longer rising time, the secondary ionization wave appears, and the energy is also deposited in the entire channel but are mainly in a local region behind the secondary ionization wave because of higher electric field as predicted in figure 1.

The lower panel of figure 2 shows the temporal evolution of energy at different rising slopes. It is interesting to find that, in cases of shorter voltage rising time, a very high voltage can be achieved when the streamer is initiated. This phenomenon has already been observed in a plane-to-plane configuration [29]. Although the geometry configuration of an SDBD is rather different with the plane-to-plane configuration studied in reference [29], similar trends are obtained and the mechanism are similar: on the one hand the fast increasing voltage leads to exponential multiplication of electrons before they are 'pushed away' from the high field exposed electrode, on the other hand the cathode sheath is not affecting the inception voltage of an SDBD significantly due to the existence of dielectric and triple point.

It is also interesting to find that, with a fixed peak voltage, the deposited total energy increase with the shorter pulse front until $t_{\text{rise}} = 1$ ns, and decrease with longer pulse rising time. In the moment of the secondary ionization wave appearance ($T \leq 250 \mu\text{s}$), the energy deposition is more or less the same. This phenomena can be explained starting from the viewpoint of an analytical solution.

The total deposited energy of SDBD was formulated in [20] as a function of permittivity, voltage, discharge length, dielectric thickness and breakdown voltage:

$$Q \approx \frac{\epsilon V_{\text{max}}^2 l_s}{16\pi d} \left(1 - \frac{V_{\text{bd}}^2}{V_{\text{max}}^2} \right) \times 10^{-9} \quad (5)$$

where l_s is the discharge propagation length. In a pulse discharge, l_s is the discharge propagation length, while in SDBD driven by sinusoidal voltage waveforms, l_s is the accumulative propagation length due to successive micro-discharges [20, 21].

We rewrite equation (5) to take into account the different morphologies of the discharge (3D filaments or 2D sheets) and the voltage rising slopes in following conditions:

(1) $t_{\text{rise}} < t_d$: in this case the voltage rising slope is ultrashort (a few nanoseconds), the discharge is in the form a quasi-uniform 2D discharge sheet. In this case we consider the discharge as a single 2D sheet, $l_s = 2h_d \nu_{ic} V_{\text{max}} / (\mu E_c^2)$.

(2) $t_{\text{rise}} \gg t_d$: we consider $t_{\text{rise}} > 10t_d$, in this case the voltage rising slope is slow, there can be multiple discharges appear simultaneously in the form of 3D filaments, in this case $l_s = l_{s2D}(5\epsilon + 4)\log(1 + 2d/h_d)/(6(\epsilon + 1)(\epsilon + 2))$, is the propagation distance of a single discharge layer of separated 3D filaments.

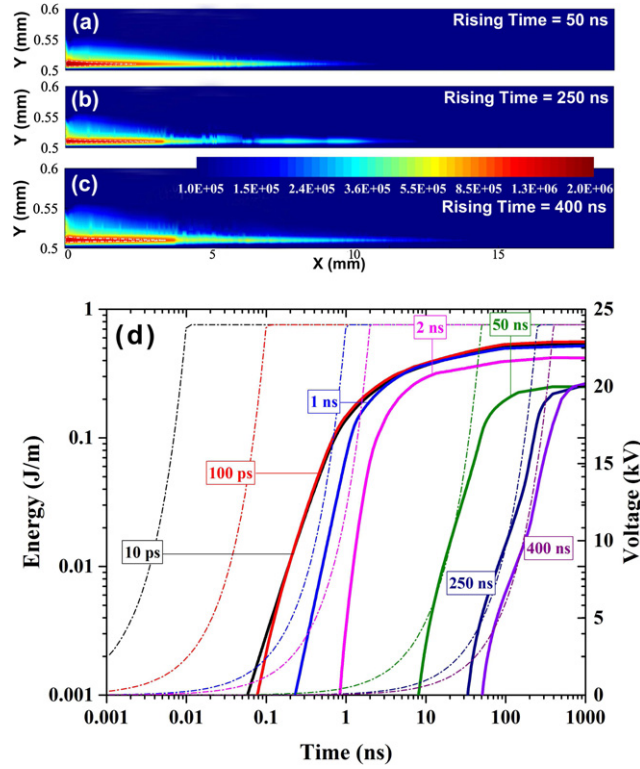


Figure 2. The spatial distribution of the total deposited energy at different rising times (a) $t_{\text{rise}} = 10$ ps (b) $t_{\text{rise}} = 250$ ns (c) $t_{\text{rise}} = 400$ ns and (d) the temporal evolution of total energy. The dash-dot lines are the applied voltage and the solid lines indicate the energy deposition from 1 ps to 1 μ s.

(3) $t_d < t_{\text{rise}} < 10t_d$: it is a transition stage when the rising time and discharge propagation time are comparable. In this case, both conditions are possible: there is only one discharge before the maximum voltage in the style of a 2D sheet, or just like in the case of condition (1), it is hard to have a definite formula for this condition, thus we assume the energy are decreasing linearly with the increasing t_{rise} .

The results of the theoretical estimation of total deposited energy are plot in figure 3(a) with measurements from independent groups of experiment and simulations. It is clearly seen that there is a significant rise of deposited energy when t_{rise} is reduced to the value comparable to t_d , agreeing well with both simulations and experiments.

4. The electro-hydrodynamic force generation

In a surface discharge, the electro-hydrodynamic force is caused mainly by the motion of negative charged particles O^- and O_2^- [30]. An analytical expression of the time averaged thrust generated by a sinusoidal arc voltage driven surface discharge has been formulated in [20], which considers the accumulative charge by the micro-discharges series occur in one sinusoidal period, and applies only to the sinusoidal voltage waveform. When the voltage rising time is decreased to the streamer propagation time scale (tens of nanoseconds), the successive micro-discharges disappears and an uniform discharge happens. We rewrite above formula as a function of t_{rise} to make it more general. Two parameters are defined here for following discussions, the time interval between micro-discharges $t_{\text{md}} = 180[V] \times t_{\text{rise}}/V_{\text{max}}$ and the relaxation time of the negative charge in the surface plasma channel $t_{\text{res}} = l_s/V_i$ ($V_i \approx 100$ m s $^{-1}$ is the ion drift velocity).

(1) $t_{\text{md}} < t_d$, the time interval between micro-discharges is smaller than the discharge propagation time, that is, the voltage rising time is short enough that only one discharge occurs, this is often the case of nanosecond discharges. Assuming the pulse duration time is two times the voltage rising time, the average force in one pulse F_{pulse} is:

$$F_{\text{pulse}} = 2.4 \times \frac{1}{t_{\text{rise}}d} \left(1 - \exp\left(-\frac{2t_{\text{rise}}}{t_{\text{res}}}\right) \right) l_{s2D}^4 \quad (6)$$

(2) $t_{\text{res}} < t_{\text{rise}}$: the voltage rising time is long enough for the negative charge to be relaxed in the channel, thus successive micro-discharges occur in one duty cycle, this is often the case of sinusoidal SDBDs. The

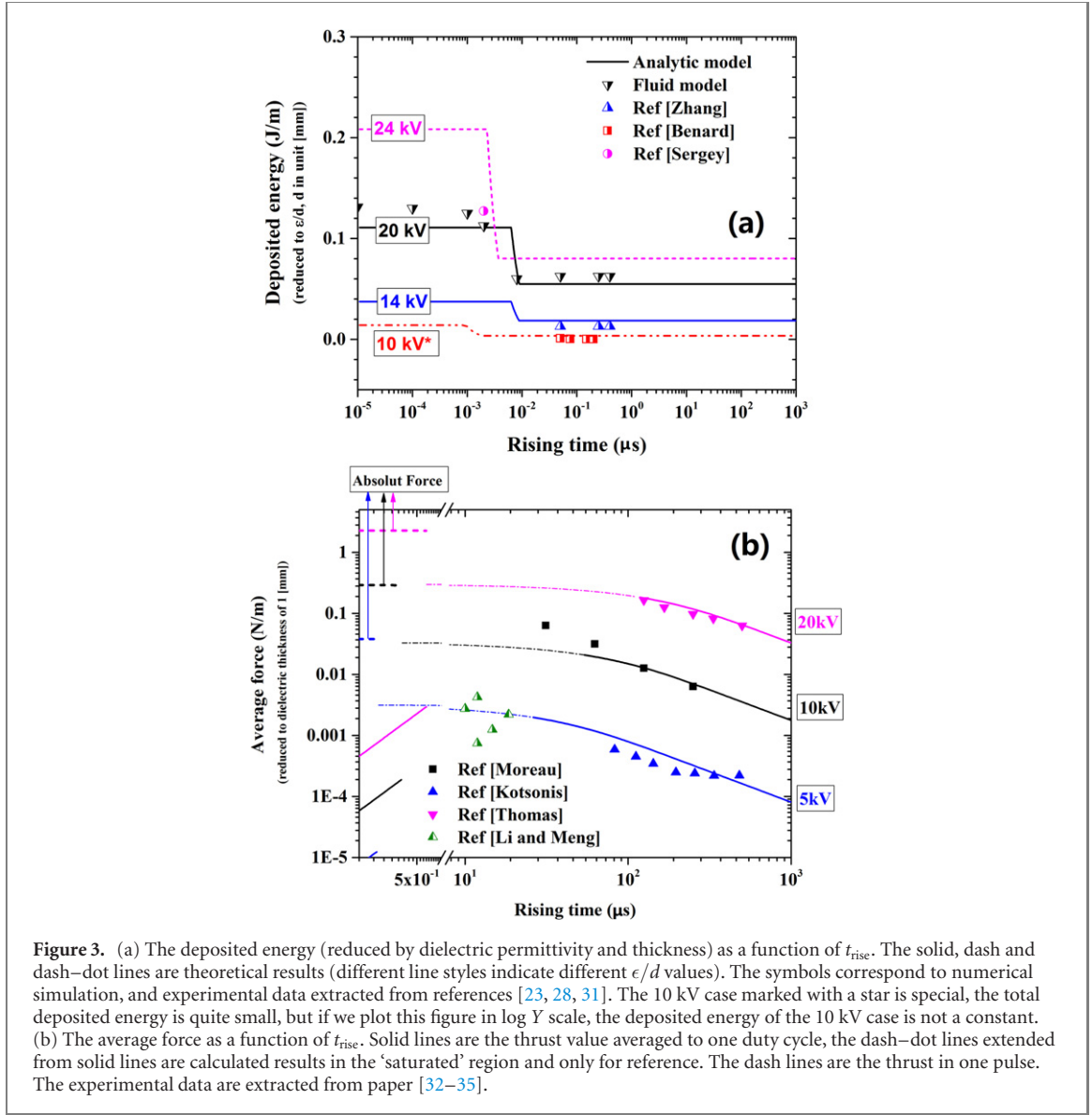


Figure 3. (a) The deposited energy (reduced by dielectric permittivity and thickness) as a function of t_{rise} . The solid, dash and dash-dot lines are theoretical results (different line styles indicate different ϵ/d values). The symbols correspond to numerical simulation, and experimental data extracted from references [23, 28, 31]. The 10 kV case marked with a star is special, the total deposited energy is quite small, but if we plot this figure in log Y scale, the deposited energy of the 10 kV case is not a constant. (b) The average force as a function of t_{rise} . Solid lines are the thrust value averaged to one duty cycle, the dash-dot lines extended from solid lines are calculated results in the ‘saturated’ region and only for reference. The dash lines are the thrust in one pulse. The experimental data are extracted from paper [32–35].

average force in one period F_{pulse} is:

$$F_{ac} = 2.4 \times \frac{1}{t_{rise}d} \left(1 - \exp \left(-\frac{t_{rise}}{t_{res}} \right) \right) l_s^4 \quad (7)$$

where $l_s = 1.5 \times 10^{-4}(1 + (9V_{max}/4dV_c)(1 - 7dV_c/6V_{max}))$, $dV_c = 600[V]$ is the normal cathode voltage fall. If $t_{res} > t_{rise}$, the discharge is saturated, negative charges cannot be fully relaxed and the discharge turn to be filamentary, equation (7) may fail.

The calculated electro-hydrodynamic force is shown together with the measurement by different groups worldwide in figure 3(b). The solid lines are the calculated period average thrust, the dash-dot lines mark the region where the discharge become saturated, while the dash lines indicate the pulse averaged thrust. The calculation and the experiment are in good agreement.

5. The characteristic map

The combination of existing experimental results, the numerical simulations and the analytical theory gives an excellent description of the behaviors of SIWs in atmospheric pressure, and allow us to draw a general two-dimensional map characterizing the reduced electro-hydrodynamics force and total energy deposition as a function of voltage amplitude and rising time.

We plot the characteristic map in figure 4. The map is an overlapping of the thrust map (the two ‘watermelon’-like petals with the yellow outlines) and the energy map (the background). Increasing the voltage, in general, leads to higher thrust and energy deposition. When the voltage rising time is reduced to

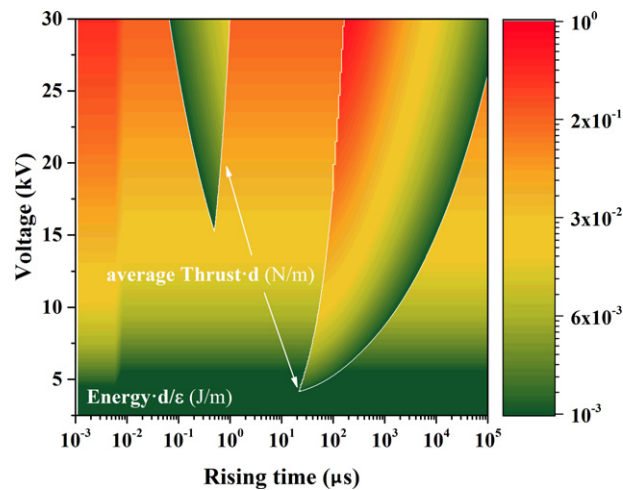


Figure 4. The characteristic map of total deposited energy and thrust in a SDBD in atmospheric pressure with t_{rise} ranging from 1 ns to 0.1 s. The maps of deposited energy and thrust are overlapped, the energy (reduced by dielectric thickness in mm and permittivity) map is the background and the two ‘watermelon’-like petals are the map of average thrust (reduced by dielectric thickness in mm).

the scale of SIW propagation time (a few to tens of nanoseconds), the deposited energy will increase significantly. To achieve higher average thrust, one has to move to the red region of the ‘watermelon’-like petals, which is also the ‘sweetest’ region in a real watermelon.

6. Conclusions

We have found that the requirements of the formation of the secondary SIW is the decay of electron density below $3 \sim 5 \times 10^{19} \text{ m}^{-3}$ while the voltage pulse is still rising. The reason is that the field in the streamer channel behind the ionization head cannot shield the still rising external electric field originates from the electrode. An analytical expression is proposed to decide the transition moment. A 2D model coupling Poisson’s equation and plasma drift–diffusion equations at atmospheric pressure can reproduce strikingly well the features of secondary SIW observed during discharge breakdown driven by sub-nanosecond pulses at atmospheric pressure.

The energy deposition and electro-hydrodynamic force are studied combining experiments, the 2D model and an extended analytical model. Calculations performed for different voltage rising times and amplitude show good agreement with the experimental results. The secondary SIW results in the spatial redistribution of the deposited energy but does not affect the total energy deposition. Decreasing voltage rising time below the characteristic discharge propagation time leads to a sharp increase of total energy deposition.

The combination of experiments, numerical simulations and analytical theories allow us a deeper insight into the physics of SIWs. A general characteristic map was drawn, providing a direct and clear description of the performance of SIWs, this map links the theoretical work and applications, and opens the door towards the target-directed design of the surface discharges.

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