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Inception of a streamer discharge in air and CO₂

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Inception of a streamer discharge in air and CO₂

ELEMENTARY PROCESSES IN GAS DISCHARGES

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July 4, 2019



Abstract

This report investigates differences between the inception of a streamer discharge in air and in CO₂. The influence of pressure and streamer polarity on inception voltage, streamer initiation delay time and characteristics of subsequent discharges during a single high voltage pulse will be studied. A pin to plate setup is used during the measurements. A high voltage pulse is applied to the upper electrode, whereas the lower one is grounded. The potential difference creates an electric field, which induces an streamer discharge. The streamer initiation is subsequently measured with a photo-multiplier.

The results show that CO₂ requires a higher voltage than air in order to initiate a streamer discharge. Furthermore, negative streamers initiate at a lower voltage than positive ones in CO₂, while in air it is the other way around. Results for delay time show that streamers initiate faster in air for both polarities. The polarity has only in CO₂ a significant influence, where the positive streamers initiate two order of magnitude faster than the negative ones. The measurements on the number of discharges show a large difference, especially at pressures near 1 bar. For both polarities the number of discharges in air are significantly higher. The differences between polarities show again opposite results between air and CO₂. Finally, it is observed that the first discharge initiates significantly faster than subsequent ones in air for both polarities. On the other hand, for negative streamers in CO₂ the differences are a lot smaller. The second discharge for the positive polarity initiates even faster than the first.

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Chapter 1

Introduction

Over the last couple of decades there were a lot of space programs started for all kinds of missions in space. Many of these programs were meant to get a better understanding of the planets in our solar system. As of 2018 there were almost fifty missions conducted to Venus [6]. A lot of research is done on the phenomena occurring in its atmosphere. It is observed by the Soviet Venera 9 and 10 that the clouds of Venus are capable of producing lightning, they obtained optical and electromagnetic evidence [7]. However, these findings are of course not enough in order to investigate the mechanisms behind the lightning in detail. This report investigates the inception of lightning in carbondioxide, the gas which Venus' atmosphere is mainly composed of. Moreover a comparison is made to air, since mechanisms of lightning on earth are better known.

Streamers are ionized regions which rapidly grow in a finger-like shape in gas, liquid or solid medium. They can be used in a variety of applications, such as ozone generation or gas and water cleaning. The process of ozone generation is performed by simply applying a streamer in air, creating first a O^* radical and then ozone [8]. Streamers can also be used to remove pollutants, such as bacteria's, tars, fine particles NO_x and SO_x from gas or water. The radicals produced by a streamer discharge induce a chemical reaction, which results in a decrease of the concentration of pollutants [9].

This report will investigate the difference in streamer inception behaviour between air and CO_2 . In particular; the inception voltage, the delay time and the characteristics of multiple discharges for streamers of both polarities will be studied. Earlier research on streamer inception in air were done by T.M.P. Briels *et al* [10]. They performed measurements with positive as well as negative streamers and found that positive discharges emerge at much lower voltage values. A. Chvyreva *et al* [11] investigated the relation between pressure and inception voltage in various gases. They found that the inception voltage scales approximately linear with pressure. Further research on the delay time was performed by M. Laan and P. Paris [12], they observed that the delay time decreases with the voltage until it reached a minimum value. After reaching this value, it increased a little bit and subsequently levelled out.

Firstly the theory behind the initiation of a streamer discharge will be given in chapter 2. Secondly, the experimental setup and method will be explained in detail in chapter 3. Thereafter in chapter 4, the results obtained from the experiments will be given, discussed and clarified with the theoretical physics behind it. Finally, conclusions of the experiments will be given in chapter 5.

Chapter 2

Theory

2.1 Initiation of a streamer discharge

A streamer discharge is a growing ionized region which can be formed in areas where the electric field is sufficiently high. It is categorized as a type of transient electrical discharge, which is also known as a filamentary discharge. The following paragraphs will describe the physical mechanisms behind the initiation of a streamer discharge.

2.1.1 Avalanche to streamer transition

A strong electric field can be formed between two electrodes when a high voltage is applied to one of them and the other one is grounded. A streamer discharge can now occur in the region between the electrodes. The initiation of a streamer discharge often starts with the ionization of a neutral atom or molecule, due to cosmic radiation or radioactive decay, creating a ion and a free electron.

The electron will be accelerated much more than the ion by the applied electric field, since it has a much higher charge to mass ratio. If an electron has gained enough energy from the electric field and collides with an atom, it can knock out another electron. This electron is in its turn accelerated and when it collides with other atoms, further ion-electrons pairs are created. These electrons are then again accelerated and can strike other atoms, creating more and more ion-electrons pairs. This chain reaction is called an electron avalanche and is visualized in figure 2.1.

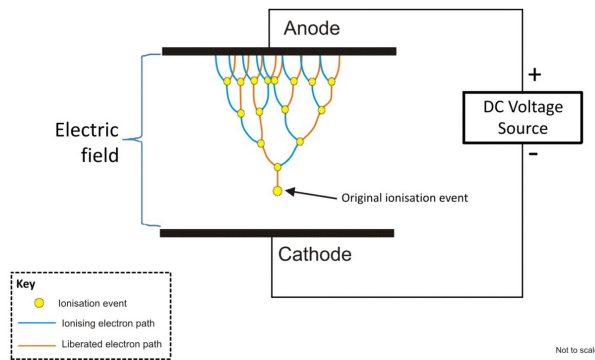


Figure 2.1: A visual representation of an electron avalanche [1].

The electron avalanches leave behind positive ions, so in time more and more space charge builds up. When it reaches a critical value, such that the intensity of the space charge field is close to that of the applied field, a streamer can be initiated [12]. This condition is also known as the Raether-Meek criterion, which will be further elaborated on in paragraph 2.1.2.

2.1.2 Raether-Meek criterion

The physics behind the initiation of a streamer discharge which are explained in the previous paragraph, will now be given in a more mathematical form. As already said, a streamer can be initiated when the space charge reaches a critical value. This critical value has been quantified by Meek. His criterion states that a streamer will be formed when the positive ion space charge field close to the anode is equal to K times the external field [13]. The value of K can be obtained by evaluating the following integral [14]:

$$\int_0^x \alpha_{eff}(z) dz, \quad (2.1)$$

where x is the distance which the avalanche can travel. α_{eff} is the effective ionization coefficient and is given by

$$\begin{cases} \alpha - \eta & \text{if } \alpha > \eta \\ 0 & \text{otherwise,} \end{cases} \quad (2.2)$$

where α represents the Townsend first ionization coefficient and η the attachment coefficient. Integration of equation (2.1) is normally performed along the direction of a electrical field line. In homogeneous fields the integral can be written in the following form [15] [16]:

$$K = \ln\left(\frac{12\pi\epsilon_0}{q_e} \frac{D_e}{\mu_e}\right) + \ln\left(\frac{x}{x_0}\right). \quad (2.3)$$

Here ϵ_0 is the dielectric permittivity in vacuum, q_e is the charge of an electron, D_e is the the diffusion coefficient, μ_e is the mobility and $x_0 = 1\text{cm}$. The ratio $\frac{D_e}{\mu_e}$ represents the characteristic energy of an electron in the avalanche. Filling in the values of the constants in equation (2.3) gives

$$K = K_0 + \ln\left(\frac{x}{x_0}\right), \quad (2.4)$$

where

$$K_0 = \ln\left(\frac{12\pi\epsilon_0 D_e}{q_e \mu_e}\right) \approx 19 \quad (2.5)$$

for a typical value of 1eV for the characteristic electron energy in avalanches. For laboratory scale experiments a typical value for x is around 1cm, therefore $\ln(\frac{x}{x_0})$ is negligible small, such that $K \approx K_0$.

2.1.3 Volume-time criterion

A streamer discharge can initiate when the applied electric field is sufficiently large. A minimum voltage must therefore be applied to one of the electrodes, which is called the inception voltage. Since the voltage is applied in the form of a pulse wave, it takes a certain amount of time until the inception voltage is reached. This type of time lag t_0 is called the boosting time lag, as shown in figure 2.2. However, when the applied voltage reaches the inception value, the streamer does not initiate immediately. This is caused by the influence of the statistical time delay t_s [17], which originates from the critical volume theory of streamers: only electrons which are liberated in a critical volume can become effective electrons which can initiate a streamer discharge. The time which this process takes is equal to the statistical delay time, which is also one of the main causes of streamer inception dispersity [2].

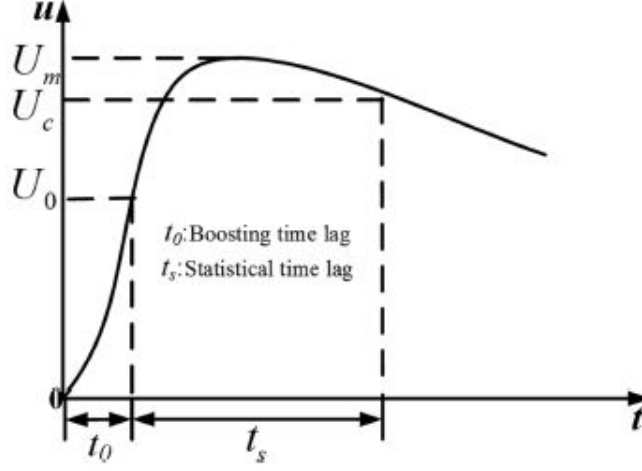


Figure 2.2: A graphical representation of the boosting time lag t_0 and the statistical time lag t_s in a streamer inception caused by a voltage pulse [2].

Statistical time lag

The volume-time criterion gives the probability of the generation of an electron with sufficient energy in a critical volume, such that an avalanche can be formed within t_s causing the initiation of a streamer discharge. For non-uniform electric fields, this criterion gives the distribution of the statistical time delay [18].

The local production rate of initial electrons per unit of time and unit volume in a gas of volume V can be denoted as $\frac{dn_e}{dt}$. However, not all electrons will be effective in initiating a streamer discharge, i.e. some of them may get ~~attracted by ions~~ present in the gas. In order for an electron to be effective it must be generated in a critical volume V_c , which will be explained below. The probability that an electron will be attracted and subsequently be attached by an atom or ion is given by $\frac{\eta}{\alpha}$, so the chance that the electron will remain active is given by

$$P_0 = 1 - \frac{\eta}{\alpha}. \quad (2.6)$$

The probability dP for a first critical electron avalanche which will be generated in a time interval between t and $t + dt$ is therefore given by

$$dP = (1 - P) \int_{V_c} \frac{dn_e}{dt} P_0 dV dt, \quad (2.7)$$

where $1-P$ is the probability that no critical electron avalanches have occurred beforehand. Integrating this equation and evaluating it at the statistical time lag t_s gives

$$P(t_s) = 1 - \exp \left[- \int_{t_0}^{t_s} \left(\int_{V_c} \frac{dn_e}{dt} P_0 dV \right) dt \right]. \quad (2.8)$$

The following part will deal with the critical volume V_c in more detail.

Critical volume theorem

In cases where the electric field is non uniform and the medium consists of an electronegative gas such as CO_2 , the initiating electrons must be liberated within a critical volume V_c to be able to start a critical electron avalanche. The criterion of a critical avalanche is when it reaches a certain size which ensures that a transition to a streamer is possible. The critical volume is a function of the distribution of the electric field and thus varies with time when a voltage pulse is applied. Its volume is bounded by two surfaces:

- i The inner surface limits the size of the electron avalanche to exceed a critical value, or in other words, the avalanche must grow over a certain minimum distance in order to subsequently form a streamer.
- ii The outer surface defines the zone where an electron avalanche growth would be possible. That is when the effective ionization coefficient α_{eff} from formula 2.2 is non-zero, so the ionization coefficient α must exceed the attachment coefficient η . Therefore the electric field within this zone must exceed the limiting value of $E_{crit} = 8.9p$ kV/mm, where p is the gas pressure [18].

2.2 Positive and negative streamers

Streamers can be split into two categories: positive and negative ones. Whether a streamer is positive or negative is determined by the polarity of the voltage applied to the electrode. Negative streamers propagate in the same direction as the electron drift, or in other words, against the direction of the electric field by accelerating their own electrons. The ‘head’ of the streamer contains an abundance of electrons, creating a negative space charge. Positive streamers on the other hand propagate in the same direction as the electric field, so against the electron drift. The streamer propagates by absorbing electrons into its positively charged head. Positive streamers require a constant supply of electrons avalanching to its head for further propagation, whereas negative streamers only need electrons for the inception. These mechanisms are illustrated in figure 2.3.

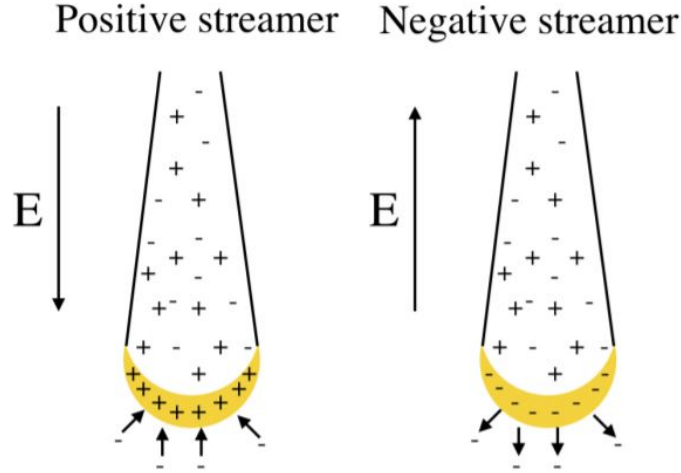


Figure 2.3: An illustration of downwards propagating positive and negative streamers. The head of the streamers is indicated in yellow, with the plus symbols representing the positive ions and the minus symbols representing the negative ions or free electrons [3].

A result of the different propagation mechanisms is that negative streamers require a higher voltage. The positively charged ‘head’ of positive streamers enhances the electrical field, thus accelerating electrons towards it, whereas with negative streamers the electrons are repelled outwards, causing a lower electric field [19] [20].

2.3 Gas properties

There are four gas properties which are important for streamer discharges: ionization energy, electronegativity, vibrational, rotational and electronic excitational levels and some other reactions like chemical ones. The following few paragraphs will give a general explanation of each property, whereas the specific properties of the used gasses will be given in the following chapter.

2.3.1 Ionization

Free electrons need to be present for a streamer to initiate and propagate, as explained in the previous paragraphs. The first free electron is often produced by cosmic radiation or radioactive decay, after which more electrons can arise from ionization of gas molecules. Ionization occurs mainly due to two mechanisms: photo-ionization and electron-impact. The electron-impact ionization is for the CO_2 gas the main source of charge generation:



with an ionization energy of $\geq 13.7\text{eV}$. For air these reactions are given by



with an ionization energy of $\geq 15.6\text{eV}$ and



with an ionization energy of $\geq 12.1\text{eV}$. Moreover, for air ionization is not only caused by electron-impact, but also by photo-ionization. It occurs when an excited nitrogen molecule emits a UV photon in the 98 to 102.5nm range, which then ionizes an oxygen molecule:



This effect is also important for positive streamer propagation. The photo-ionization is a non-local effect, since emitted photons from the nitrogen atom can ionize an oxygen atom at some distance away. Excited nitrogen atoms in the streamer's head can therefore ionize oxygen molecules just in front, thus maintaining a constant electron flow to the head. For negative streamers on the other hand, photo-ionization plays a negligible role. The electron impact ionization effect is dominant, since their velocity is comparable to the drift velocity of electrons in the local field [21].

2.3.2 Electronegativity

Electronegativity is a chemical property which describes the tendency of an atom or molecule to attract electrons. This property therefore influences how many free electrons are lost due to attachment to other molecules. The electronegativity of an atom is determined by both its atomic number and the distance between the charged nucleus and the valance electrons. The more electronegative an atom or molecule is, the more it attracts electrons. The opposite is electropositivity which is a measure of the ability of an atom or molecule to donate electrons. The value of the electronegativity can vary with its chemical environment, but usually it is considered to be a transferable property, so that the same value is applicable in a variety of situations [22].

2.3.3 Vibrational, rotational and electronic excitational levels

During the electron avalanche and propagation phase there are many collisions of electrons with ions. How much energy an electron loses at an inelastic collision is determined by the vibrational, rotational and electronic excitational levels. This loss of energy has as a consequence that the electron's energy can drop under a certain level, making it unable to ionize other gas molecules.

Two types of gas can be distinguished: atomic and molecular gas. The particles of a molecular gas consists of multiple atoms, which allow vibrations and rotations of the molecule, resulting in that a molecular gas can have many vibrational, rotational and excitational levels, whereas an atomic gas only has a few excitational levels. In the experiments however, only molecular gas will be used.

2.3.4 Other reactions

There are a couple of other reactions which have an influences on the system, like chemical and recombination reactions. Properties such as the number of ions and electrons, exited states and dissociation are effected by these reactions. The number of these reactions could become very large when considering a molecular gas at a relative high pressure. Earlier literature constructed a quantitative model of the chemical dynamics of the streamer head and including 80+ species accounting for 800+ reactions [23].

Chapter 3

Experimental setup and method

The experimental setup and method which are used during this experiment will be described in detail in paragraph 3.1. Furthermore, the observation of the signals will be explained in paragraph 3.2. Finally, all the tested parameters with their influence on the system will be elaborated on in paragraph 3.3.

3.1 Setup and method

All experiments were performed using the setup depicted in figure 3.1. This paragraph will explain the workings of the whole setup and the functionality of each individual component in detail.

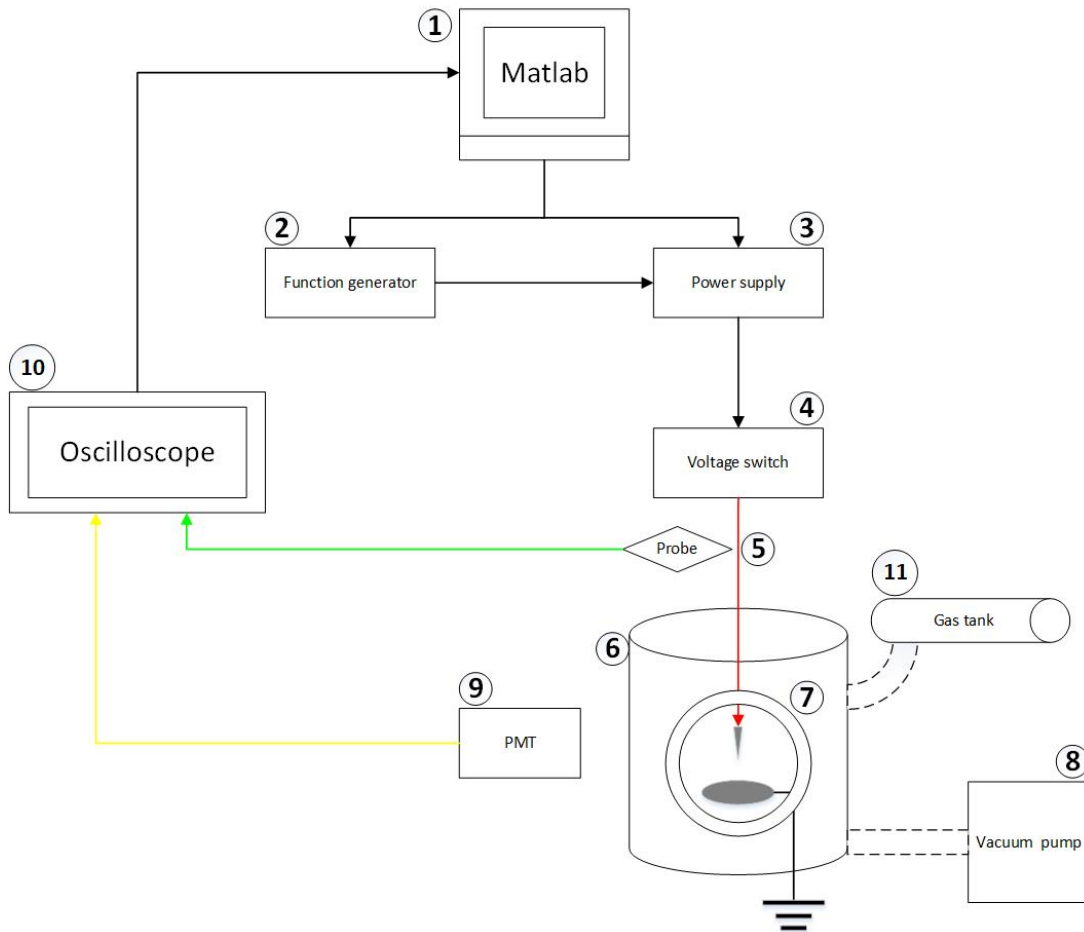


Figure 3.1: A schematic overview of the experimental setup for streamer discharge generation and measurement.

- ① PC: the PC runs Matlab 2017a which controls the whole system. Matlab itself runs the application AppFinder (written by Shariar Mirpour), which is used to control the function generator ②, the HV power supply ③. It also reads out and saves the signal obtained from the oscilloscope ⑩. The program can set the values of the pulse duration, pulse frequency, voltage value and the number of acquisitions.
- ② Function generator: the function generator is controlled by the PC ①. It generates a rectangular pulse function and sends it then to ~~the HV power supply ③.~~
- ③ High voltage power supply: the Spellman SL150 HV power supply provides the voltage for the system and is also controlled by the PC ①. The voltage values it can generate ranges from 0 to 30kV.
- ④ Voltage switch: the voltage switch ~~can change the polarity of the system.~~
- ⑤ Voltage probe: The Northstar PVM-1 voltage probe measures the voltage which is applied to pin to plate setup ⑦ and converts it to a signal which is send ~~a~~ to the oscilloscope ⑩ through channel 1.
- ⑥ Vessel: inside the vessel the pin to plate setup ⑦ is located as can be seen in figure 3.2. The vessel is connected to the gas tank ⑪, such that it can be filled with various gasses. Furthermore the vessel is connected to the the vacuum pump ⑧, which can lower the pressure inside to a minimum of 5mbar.

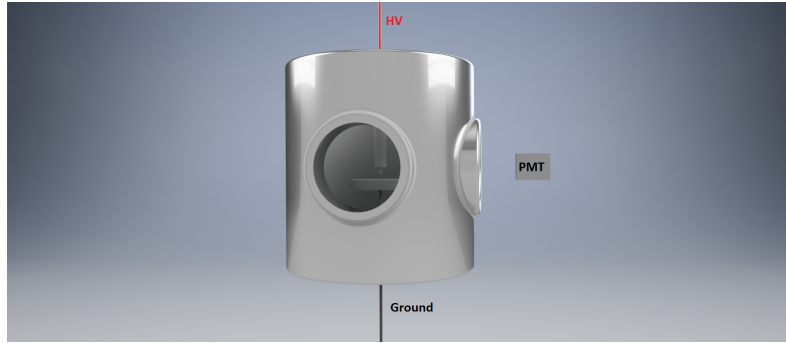


Figure 3.2: A more detailed 3D representation of the vessel with the pin to plate setup located inside and the PMT at the right.

- ⑦ Pin to plate setup: the pin to plate setup, depicted in figure 3.3, is placed inside the vessel ⑥. It consists out of a sharp needle-like upper electrode with a length of 1cm and a round lower electrode, with a spacing of 3cm in between. A clarification of this distance is chosen, will be given in the next chapter in paragraph 3.3.6. The voltage pulses generated by ② and ③ will be applied to the upper electrode, whereas the lower electrode is grounded.

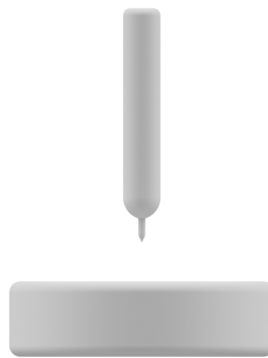


Figure 3.3: A more detailed representation of the pin to plate setup. With the upper electrode connected to the HV and the lower to the ground.

- ⑧ Vacuum pump: the vacuum pump is connected to the vessel ⑥ in order to change the pressure inside. The pump can set a range of pressures between 0.005bar and 1bar.
- ⑨ Photo multiplier (PMT): the photo multiplier is used to observe the initiation of streamers. The PMT converts the photons which are generated by a streamer to electrons and sends the signal through channel 4 to the oscilloscope ⑩. Its sensitivity can manually be set and determines the factor at which the signal is amplified. A more detailed elaboration of its exact workings will be given in the following paragraph.
- ⑩ Oscilloscope: the Teledyne LeCroy oscilloscope monitors the whole system. It first collects the data from the signal received from the PMT ⑨ and the voltage probe ⑤, where after it is send back to the PC ①.
- ⑪ Gas tank: the gas tank is connected to the vessel ⑥ in order to fill it with different kind of gasses. During the experiments ambient air, dry air, a nitrogen-oxygen mixture and carbondioxide were used.

3.1.1 Working of the photo-multiplier (PMT)

A photo-multiplier or PMT is a device which is extremely sensitive to light in the ultraviolet, visible and near-infrared spectrum. These detectors multiply the current induced by an incident photon as much as a 100 million times, in subsequent dynode stages, which enables the detection of single photons even when the incident light flux is low.

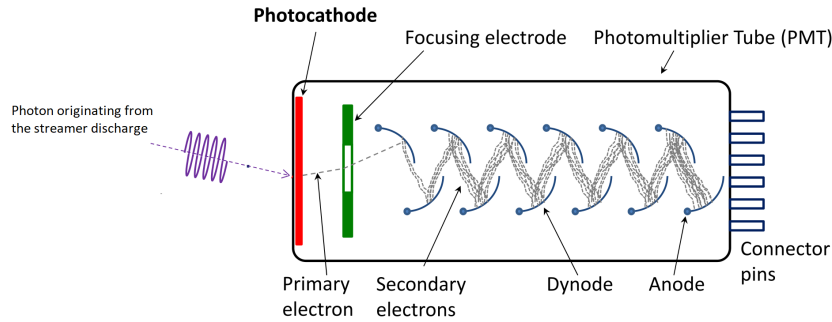


Figure 3.4: Schematic overview of a photo-multiplier with an incident photon originating from the streamer discharge [4].

A schematic overview of the inside of a photo-multiplier is displayed in figure 3.4. Incident photons released during a streamer discharge strike the photocathode, which consists usually of a vapor-deposited conducting layer. Due to the photo-electric effect an electron is generated and ejected from the surface. A focusing electrode subsequently directs the electron towards the electron multiplier, consisting of multiple dynodes, where electrons are multiplied by the process of secondary emission; when an electron has sufficient energy it can induce the emission of secondary electrons when hitting the surface of the dynode. Each subsequent dynode is held at a higher positive potential than the preceding one, in order to accelerate the incoming electrons. The low energy electrons liberated from the first dynode are thus accelerated by the second one, gaining enough energy to liberate more electrons. This process is repeated a couple of times, after which all the electrons are ‘collected’ by the anode, thus creating a sharp current pulse. This current pulse is then read out on the oscilloscope, which is explained in more detail in the next paragraph.

3.2 Signal observation

After a voltage pulse is applied to the upper electrode a streamer can be initiated. The photons which are generated during a discharge are converted into a signal by the PMT and subsequently read out by the oscilloscope. An example is displayed in figure 3.5.

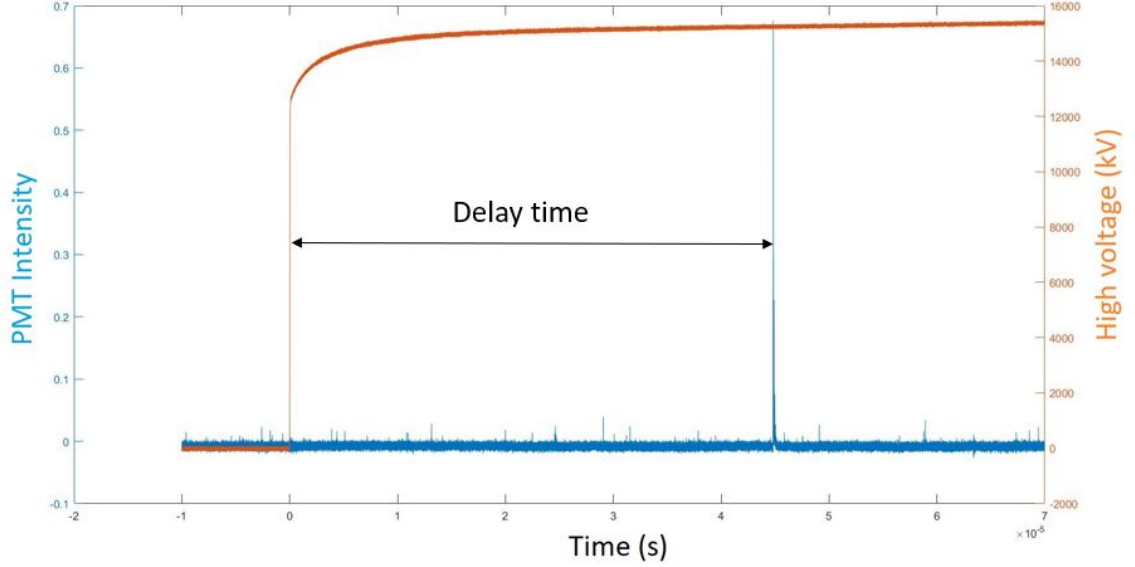


Figure 3.5: Signal output on the oscilloscope for a streamer discharge, the signal from the photo multiplier is indicated in blue and the high voltage is indicated in red.

The delay time is defined as the time interval between the high voltage rise and the streamer inception. The streamer inception can be observed as a large peak in the PMT signal, as can be seen in the figure above. The oscilloscope will classify a peak as an inception if the absolute value is above the threshold value. This value can manually be set and is chosen depending of the sensitivity upon the PMT. The threshold value is most of the time kept at 200mV, since the PMT is always set at approximately the same sensitivity. Only for measurements with CO_2 this value is adjusted, since its discharge is very faint. Finally the data including delay time, number of discharges and time interval between consecutive discharges will be send to the PC and then be saved.

3.3 Parameters

There are in total seven parameters which are involved during the experiments: type of gas, polarity and value of the voltage, pressure, pulse duration, frequency and distance between the electrodes. Three of them are fixed and initially investigated in order to find suitable values, while the other four are varied during the experiments. This can be seen in figure 3.6, which depicts a schematic overview of all the parameters with their corresponding ranges. Note that not all possibilities of combinations of parameters are drawn, however all are tested. The properties and influence of each parameter on the system will further be elaborated on in the coming paragraphs.

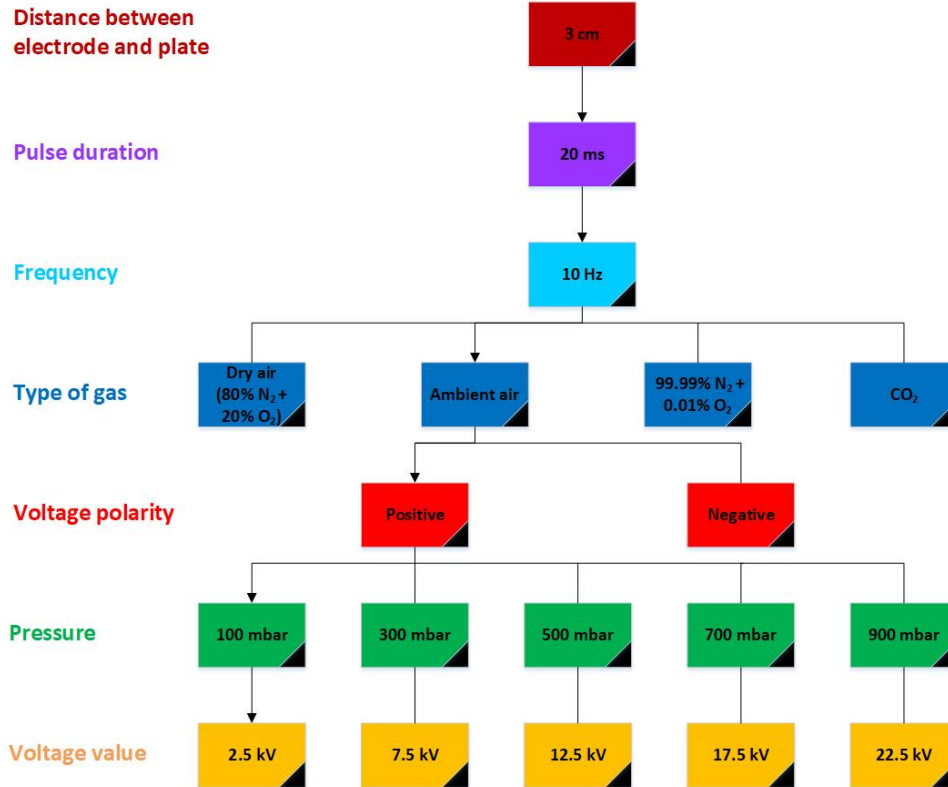


Figure 3.6: A schematic overview of all parameters involved in the experiments. Only dry air is not used during the measurements with the negative polarity.

3.3.1 Type of gas

There are four types of gas used during the experiments: ambient and dry air, a 99.99% $N_2 + 0.01\%$ O_2 mixture and CO_2 . The dry air is artificially composed and consists of N_2 and O_2 , which contribute for 80 and 20 percent respectively. Furthermore, the usage of the N_2 - O_2 is only meant to be a tool for the clarification of the differences between air and CO_2 . Since dry air is composed of multiple components, it could give a useful insight in the workings of one component only. So the delay time and number of discharges will not be investigated for this particular gas.

3.3.2 Voltage

The voltage will be varied in two different ways. First of all, both polarities will be used: positive and negative HV pulses. According to earlier research negative streamers require a higher voltage value in order to emerge than positive ones [19].

The second aspect of the voltage which will be varied, is its value. However at each pressure only one value will be used, such that the ratio $\frac{\text{pressure}}{\text{voltage}}$ will be held constant at $40 \frac{\text{mbar}}{\text{kV}}$. So its direct influence on the system will not be investigated.

3.3.3 Pressure

The pressure in the vessel determines the number of molecules which are present. It will be varied in a range between 100mbar and 900mbar, as seen in figure 3.6. Its effect on inception voltage, delay time, number of inceptions and time interval between consecutive discharges will be investigated.

The applied electric field will accelerate free electrons, which can collide with other molecules. This will slow down the electrons in their way to or from the tip of the streamer. By increasing the pressure the number of molecules present is thereby also increased, resulting in a smaller mean free path which the electrons can travel. The electrons are therefore accelerated over a smaller distance, which could result in not gaining enough energy for ionization.

Streamers need initial electrons in order to emerge. In the case when multiple discharges during a single pulse are observed, the leftover species from the first discharge could influence the subsequent ones. The presence of these leftover particles will probably result in a faster initiation of subsequent discharges, since the statistical time lag t_s is now drastically reduced.

3.3.4 Pulse duration

The pulse duration is the time length at which the HV is applied to the upper electrode. Obviously, the longer the system is exposed to the HV, the higher the chance is for a streamer to initiate and the more streamer inceptions there will be. However, the duration of the pulse will probably have no effect on the delay time.

The effect of the pulse duration won't be further investigated during the experiments. So it will be held constant at a value of 20ms, which is enough to initiate a couple of discharges. A longer pulse duration leads to an increased chance of a breakdown and thus a smaller range of suitable voltages at which can be measured.

3.3.5 Frequency

The frequency determines the repetition rate at which the HV pulse is applied. As mentioned in paragraph 3.3.3, leftover species from previous discharges can have an influence on subsequent ones. So when the time between two pulses is too short, these leftover particles can influence the following discharges. On the basis of earlier work [24] [25], the frequency is determined and set at a fixed value of 10Hz, resulting in a relaxation time of 80ms. At this value the influence of leftover species of previous discharges is negligible.

3.3.6 Distance between electrodes

The distance between the upper electrode and the lower plate influences the inception and breakdown voltage. A larger gap leads to higher inception and breakdown voltages and thus a larger range at which can be measured, since both are multiplied with the same factor. The gap is therefore fixed at a distance of 3cm. Shorter distances won't provide a sufficient large range of voltages at which can be measured, since the inception and breakdown voltage are too close together. On the other hand larger distances would give problems for measurements with higher pressures.

Chapter 4

Results and discussion

This chapter will show and discuss results obtained from the experiments. Firstly, a discussion of the inception voltage of positive as well as negative streamers in different gasses is presented. Next, results of the streamer initiation delay time of carbondioxide and air are given and a comparison is made. Finally, the characteristics of multiple discharges during one single voltage pulse are discussed. The data presented below is an average of 500 trials of streamer discharge initiation.

4.1 Inception voltage

The inception voltage is defined as the minimum voltage which is required to initiate a streamer discharge. Its characteristics are investigated for positive and negative streamers in four different types of gas. Due to circumstances measurements of negative streamers in dry air could not be performed. Figure 4.1 shows the inception voltage for positive and negative streamers in different gasses at different pressures.

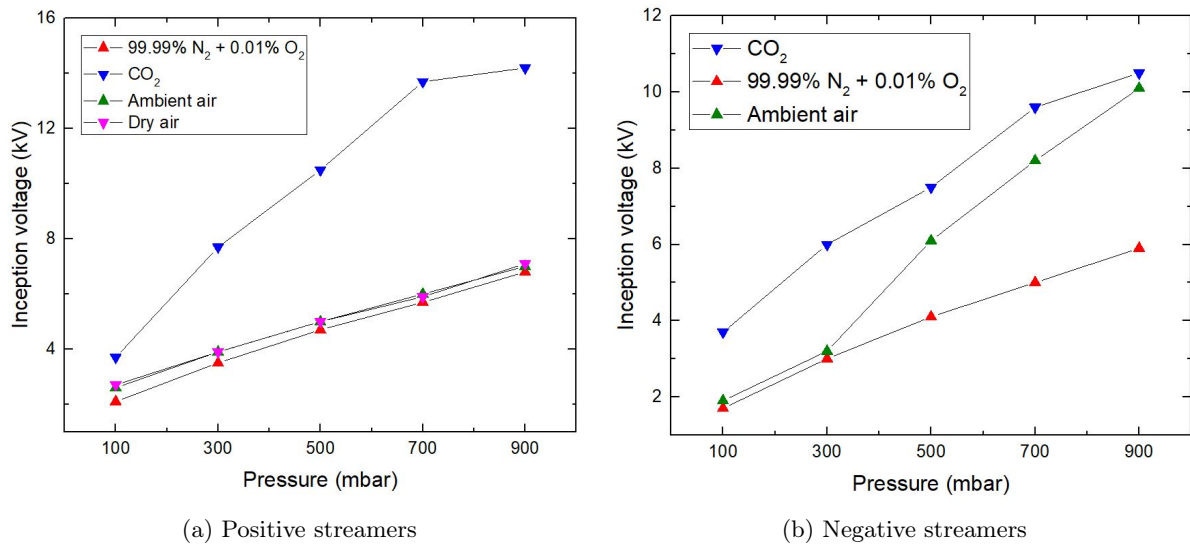


Figure 4.1: The inception voltage for the N₂-O₂ mixture, CO₂, dry and ambient air.

Both graphs show that for all gasses the inception voltage increases with pressure. The mean free path for the electrons is much higher at lower pressures, as explained in paragraph 3.3.3. Electrons are thus accelerated for a longer amount of time before colliding with another ion or atom. This allows the electrons to gain enough energy for impact ionization. Therefore lower voltages are sufficient for streamer initiation when the pressure is decreased.

It is also observed that the inception voltage for CO_2 in both cases is significantly higher than for other gasses. The differences in the ionization energy do not give sufficient clarification; despite that O_2 has a lower ionization energy than CO_2 [26] [27], N_2 has a higher one [28]. A better explanation could be given on the basis of the photo-ionization effect, which only occurs in ambient and dry air. The energy of the emitted photon from the excited nitrogen molecule ranges between 12.1eV and 12.7eV, which is lower than the ionization energy of CO_2 [29]. The electrons for impact on the nitrogen molecules do therefore not require to have the minimum ionization energy of nitrogen in order to induce ionization. As the photo-ionization effect is less effective in the electron avalanche and propagation phase for negative streamers [21], the inception voltage of ambient air is a lot closer to CO_2 . Simulations performed by the simulation of streamers group from Ute Ebert at CWI also confirm that photo-ionization less effective for negative streamer, however, why this is the case, is not clear and more research is required to get a better understanding.

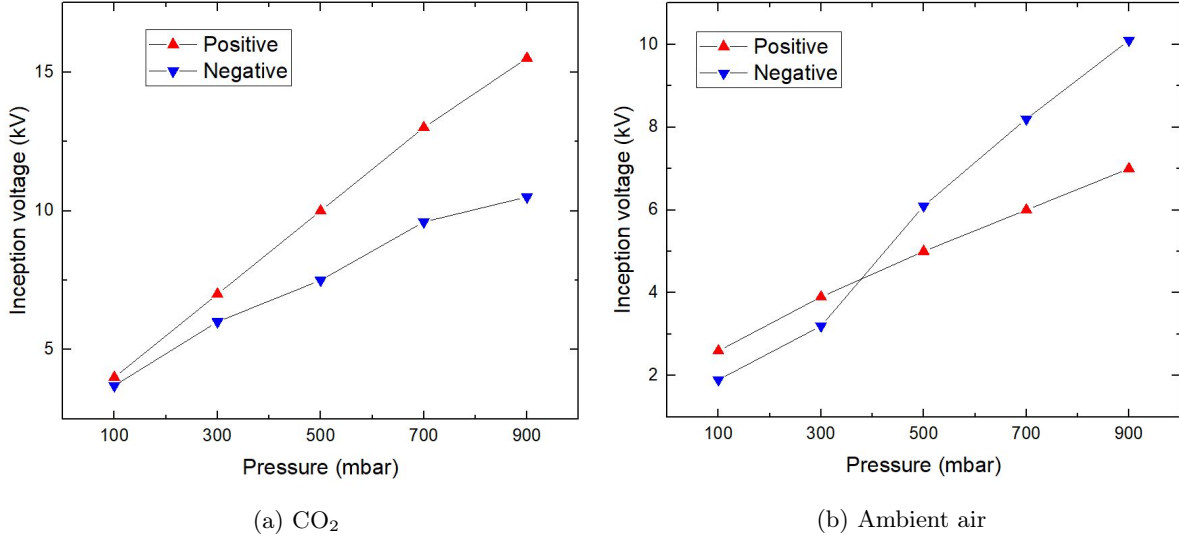


Figure 4.2: The inception voltage for positive and negative streamers.

Graph 4.2 shows the difference in inception voltage for positive and negative streamers in CO_2 and ambient air. For CO_2 the positive streamer requires a higher voltage to initiate. Whereas in the case of ambient air it is the other way around. Photo-ionization can again provide a possible answer why this is observed; whereas for positive streamer the electron production is mainly governed by the photo-ionization effect, for negative streamers the effect is negligible compared to the impact ionization effect. The positive streamer initiates easier, as ambient air relies a lot on the photo-ionization effect for electron generation. However, in CO_2 the photo-ionization effect plays no insignificant role due to the low oxygen concentration. Moreover, positive streamers are more dependent upon free electrons for initiation and subsequent propagation than negative streamers [21]. Due to the electro-negative nature of CO_2 there are less free electrons for initiation present, so a higher voltage is required for inception of positive streamers.

4.2 Streamer initiation delay time

The time interval between the start of the HV pulse and the first streamer discharge is defined as the delay time, as can be seen in figure 3.5. The delay time is measured for positive and negative streamers in air and CO₂ at different pressures. In order to obtain comparable results, the ratio $\frac{\text{pressure}}{\text{voltage}}$ is kept constant during every measurement series, with the same voltage at a certain pressure for the positive and negative polarity in both gasses. The results for positive and negative streamers in CO₂ and ambient air are shown figure 4.3.

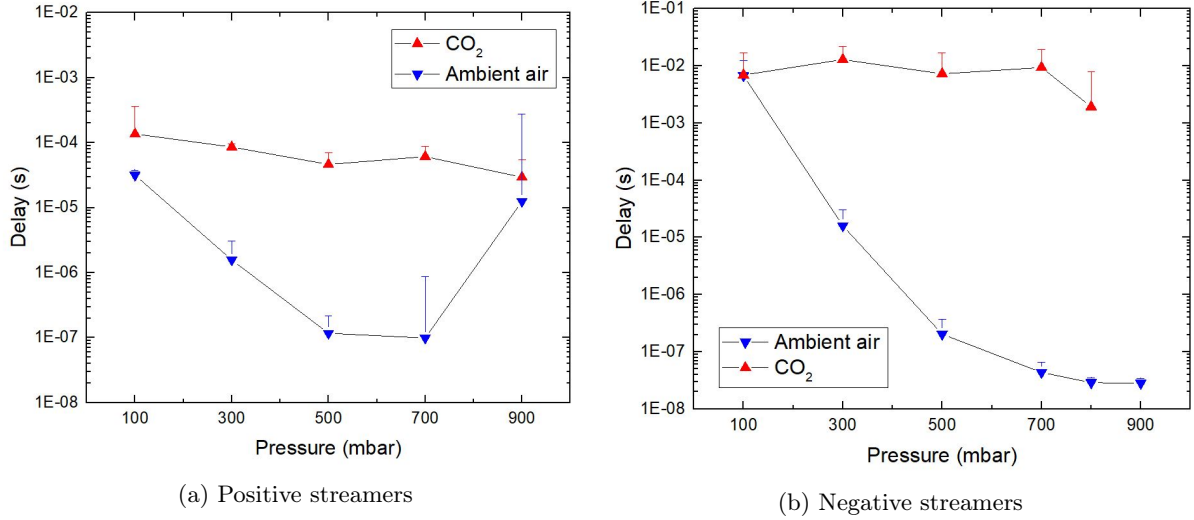


Figure 4.3: The inception delay time for CO₂ and ambient air. Error bars are only showed in the positive direction, the bars in the negative direction are not presented for visibility reasons due to the logarithmic scale.

It is seen that the delay time for positive and negative streamers in CO₂ is higher than in air. This difference could be caused by the way electrons are moved towards the critical volume. The most simple way electrons are transported towards that zone is by direct influence of the applied electric field without any significant interaction with other molecules. Another way is via attachment and detachment mechanisms. In air an electron can attach to O₂ forming O₂⁻ and in carbon dioxide to CO₂ forming CO₂⁻. These negatively charged ions are then subsequently attracted towards the ionization zone, with the O₂⁻ moving significantly faster, since CO₂ is around 1.4 times as heavy as O₂, resulting in faster initiation. Moreover the photo-ionization effect could have an influence as well: since nitrogen only has to get to an excited state to eventually induce ionization and oxygen has a lower ionization potential, the likelihood of a successful electron avalanche in air is larger than in CO₂ at a constant voltage and pressure. This results in a faster streamer initiation, or in other words, a smaller delay time.

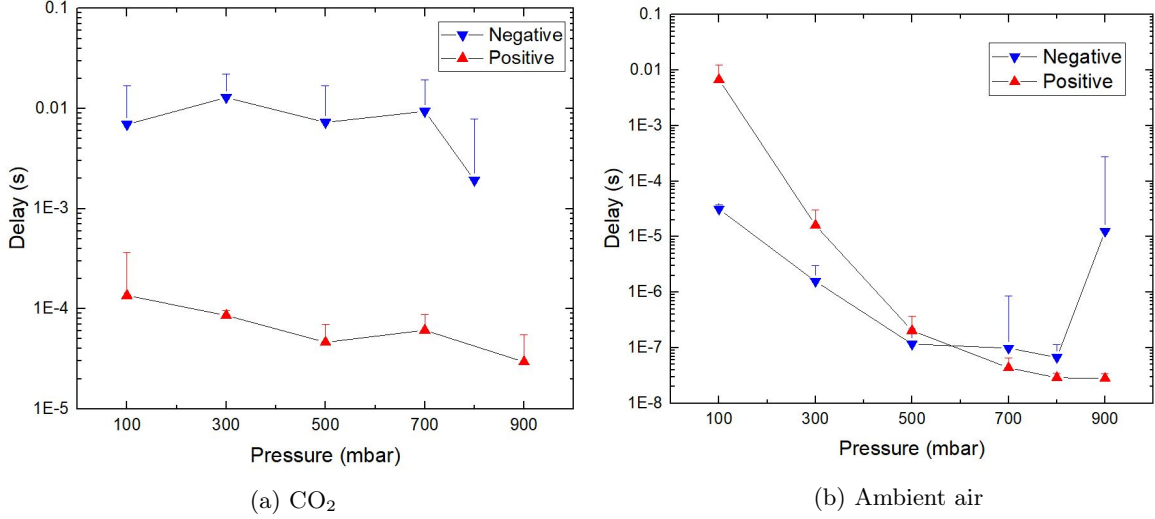


Figure 4.4: The inception delay time for positive and negative streamers. Error bars are only showed in the positive direction, the bars in the negative direction are not presented for visibility reasons due to the logarithmic scale.

The difference between positive and negative streamers is shown in figure 4.4. Positive streamers initiate approximately 100 times faster than negative ones in CO₂. When looking at the figures of the electric field around the pin, which are shown in appendix C.1, it is seen that there is a large field gradient around the tip of the needle. In the case of the negative polarity the electric field is directed inwards, so the electrons are repelled away from the pin towards a lower electric field region. Meanwhile for the positive polarity the electric field is directed outwards, so electrons are attracted inwards towards a higher electric field region. This suggests that the electrons reach faster the required energy to induce ionization for the positive polarity, resulting in a faster streamer initiation.

For both positive and negative streamers the delay time decreases with pressure, especially for air. A possible explanation can be given on the basis of the volume-time criterion. As the pressure and thus the number of gas molecules increases, the chance that an electron will be liberated in the critical volume V_c also increases, thereby decreasing the statistical time lag t_s .

4.3 Multiple discharges

The number of streamer discharge inceptions and the delay time between consecutive discharges during a single voltage pulse are measured. Just as with the measurements in the previous paragraph, the voltage used at a certain pressure for positive and negative polarity in all gasses is fixed. And the ratio $\frac{\text{pressure}}{\text{voltage}}$ is kept constant during every measurement series.

4.3.1 Number of discharges

The difference in the number of discharges between ambient air and CO₂ is shown in figure 4.5. Especially streamers in air of both polarities show a higher number of inceptions as the pressure is increased, which again can be explained on the basis of the critical volume theorem; as the number of species present is increased, the chance that electrons will be in the right place at the right time to form a critical avalanche also increases.

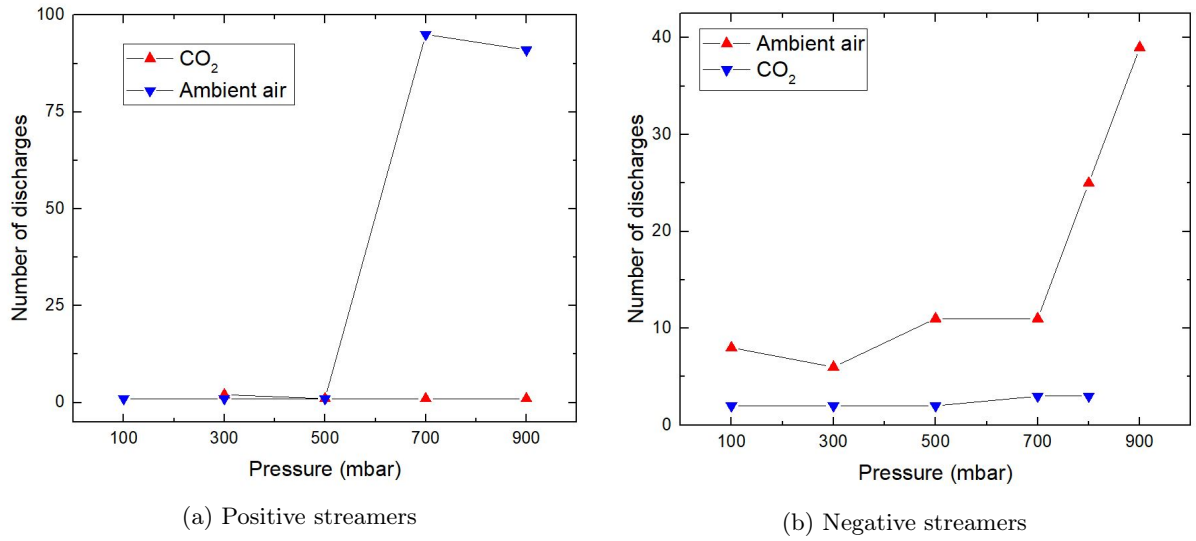


Figure 4.5: The number of discharges measured in CO₂ and ambient air.

The results for both positive and negative polarity show a large difference between ambient and CO₂, especially at relative high pressures. Due to the electro-negativity of CO₂, it bonds to more free electrons than the air molecules. Evaluating the electron attachment rate for air and CO₂ at the appropriate reduced electric field confirms this behaviour. This suggests that after a discharge less electrons are left over for the initiation of subsequent ones, in the case of CO₂. This would therefore result in a lower number of discharges. The figures of the electron attachment rate and the reduced electric field in air and CO₂ can be found in appendix C.2 and B.1 respectively.

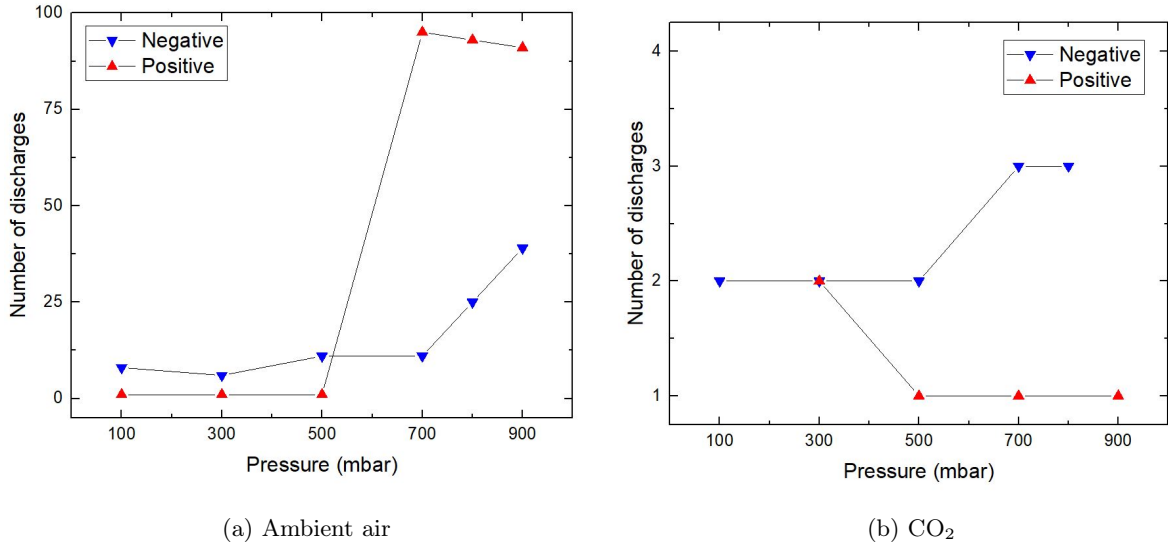


Figure 4.6: The number of discharges for positive and negative streamers.

Figure 4.6 shows the differences between streamers of positive and negative polarity. One can see that these differences are significantly higher in air than in CO₂ especially at relative high pressures. It is again observed that air shows opposite behaviour with respect to CO₂. It is expected that space charge layers which are left over from a discharge can obstruct the forming of subsequent streamers. These space charge layers could be the remains of a streamer's head. This hypothesis is further elaborated on in the next paragraph. However, more research is needed on this topic in order to give a better explanation of this observed behaviour.

4.3.2 Time interval between consecutive discharges

The time between consecutive discharges during a single voltage pulse is measured and all results are shown in the appendix A.1. Only cases where multiple discharges are observed, are displayed. In the graphs the interval between consecutive discharges is indicated in the form of $dx - y$, where x and y represent the n^{th} and $n^{\text{th}}+1$ discharge respectively.

The results of positive and negative streamers in ambient are given in the figures found in appendix A.1.1 and A.1.2 respectively. In both cases the first discharge always takes a relative low amount of time to initiate compared to all of the subsequent discharges, for which the delay time is approximately the same. The only exception is shown in graph A.1, where the second discharge takes a significant longer time than the rest. A possible reason for why this first phenomenon is observed, is related to the remaining space charge layers from the first discharge. These space charge layers may form a barrier which obstruct the forming of new streamers. Especially in regions where a lot of charge is clustered, the local electric field could have a strength comparable with the applied field. An interference with the global electric field could result in that new streamers can't form and propagate. The space charge species require some time to recombine to neutrals again, which could explain the delay time difference between the first and the other discharges.

The space charge layers which could obstruct the forming of new streamers can originate from the old streamer's head. A. Luque *et al* [20], Z. Bonaventura [30], X. Wang *et al* [31] all have investigated the electric field strength originating from the head of positive and negative streamers in atmospheric air. The parameters which are used in their setups are comparable with ones used in this report. The values of the electric field strength of the streamer heads they found, ranges from $75 \frac{\text{kV}}{\text{cm}}$ to $175 \frac{\text{kV}}{\text{cm}}$. The applied electric field evaluated at three different voltages is shown in appendix C.1. The electric field shows a large gradient around the needle tip, with maximum values of $200 \frac{\text{kV}}{\text{cm}}$. This suggests that the electric field of the head can interfere with the global field, since their values are in the same order of magnitude.

Figure A.6 shows that the delay of the first streamer discharge of negative polarity in CO_2 is shorter than the subsequent ones, just as in atmospheric air, however the differences are significantly smaller. Furthermore for streamers of positive polarity the delay of the second discharge is even shorter than the first one, as seen in figure A.5. Table B.1 in appendix B.2 shows the recombination rates of possible leftover ions after a discharge. The rates for the leftover ions in air are significantly smaller than in CO_2 , which confirms the results of the experiments; the remaining space charge layers in CO_2 will dissolve more quickly due to the higher recombination reaction rate. Therefore the difference between the time interval of the first and subsequent discharges is a lot smaller. Although not all the recombination rates of the ions in CO_2 are found, it is expected that these are also in the same order size. Moreover, the remains of the positive streamer will recombine extra quickly due to the electro-negativity of CO_2 , which could explain the difference between the positive and negative polarity.

Chapter 5

Conclusions

In this report differences between the inception of a streamer discharge in air and CO_2 were investigated. To be more specific; influence of pressure and streamer polarity on inception voltage, delay time and characteristics of multiple discharges during a single voltage pulse were studied.

The results for the inception voltage showed that a higher voltage was required for streamer initiation when the pressure was increased, which is due to the fact that the mean free path of electrons changes when the pressure is varied. It was also observed that CO_2 has a higher inception voltage than air for both polarities. And the comparison between the positive and negative polarity showed that the inception voltage is lower for negative streamers in CO_2 , whereas in air it is the other way around. The latter two observations are both caused by the photo-ionization effect. However, more research needs to be performed on why the photo-ionization is less effective during the avalanche phase for streamers of negative polarities.

Secondly, it was found that an increasing pressure resulted in a decrease of the inception delay time. An explanation was given on the basis of the volume-time criterion. Furthermore, the inception delay time of CO_2 was observed to be significantly higher than that of air. Two possible causes were posed; the first one was based on the way the electrons were transported towards the critical volume region and the second one was related to the photo-ionization effect. The influence of the polarity of the streamer was observed to be significant in CO_2 , whereas the differences in air were a lot smaller. The negative streamers in CO_2 took two orders of magnitude as long to initiate compared to positive streamers, which could be clarified by looking at the direction and the large gradient around the electrode tip of the electric field.

Thirdly, the number of discharges during a single HV voltage pulse were investigated. It was seen that an increase in pressure led to a higher number of discharges, especially in air. An explanation was given again on the basis of the volume-time criterion. Moreover, the comparison between air and CO_2 showed a large difference; the number of discharges in air were significantly higher especially at pressures close to 1 bar. The electro-negative nature of CO_2 leads to a different electron attachment rate than of air, which could explain the results. Why differences between the positive and negative streamers are observed, remains unclear and further research is needed to get a better understanding.

Finally, results of measurements of the time interval between consecutive discharges during a single voltage pulse showed that for both polarities in air the first discharge initiated a lot faster than all the subsequent ones. The electric field originating from the left over space charge layers could form a barrier, thus slowing down the subsequent initiations. Results for negative streamers in CO_2 showed similar results, however differences in delay time between the first and subsequent discharges were much smaller. A possible explanation was based on the recombination rate of the leftover species after a discharge. For streamers of the positive polarity in CO_2 the second discharge initiated even faster than the first one. Unfortunately, it was not possible to investigate the recombination process of the remaining space charge layers due to time limitations. Further research should be performed to confirm the proposed hypothesis.

To conclude, this report tried to explain all results on the basis of physical mechanisms. However, the measurement equipment did often not provide enough information in order to gain an accurate insight of what is exactly happening. Measurements with an ICCD-camera could give a useful insight with respect to the inception cloud, which may be helpful to clarify some observations.

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Appendix A

Results

A.1 Time interval between consecutive discharges

A.1.1 Positive streamers in ambient air

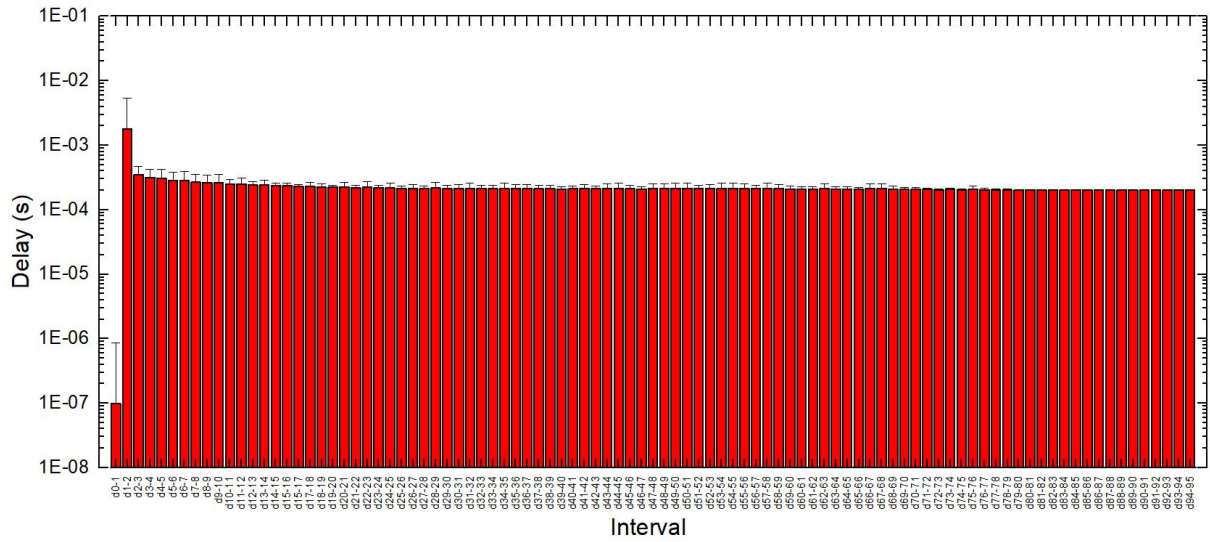


Figure A.1: Time interval between consecutive discharges during a single voltage pulse for positive streamers in ambient air. Measured with 17.5 kV at 700 mbar

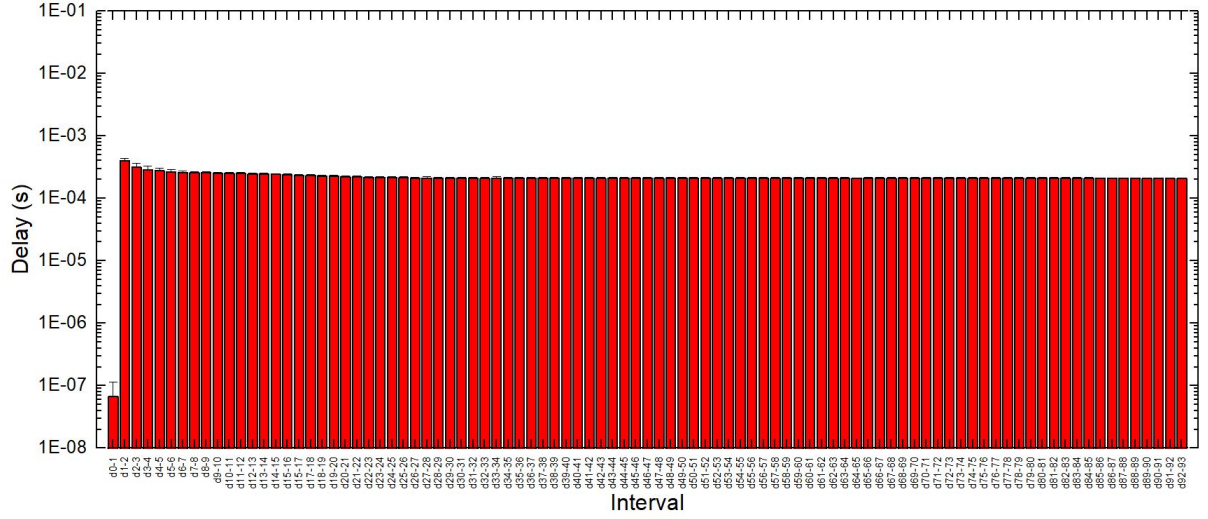


Figure A.2: Time interval between consecutive discharges during a single voltage pulse for positive streamers in ambient air. Measured with 20 kV at 800 mbar

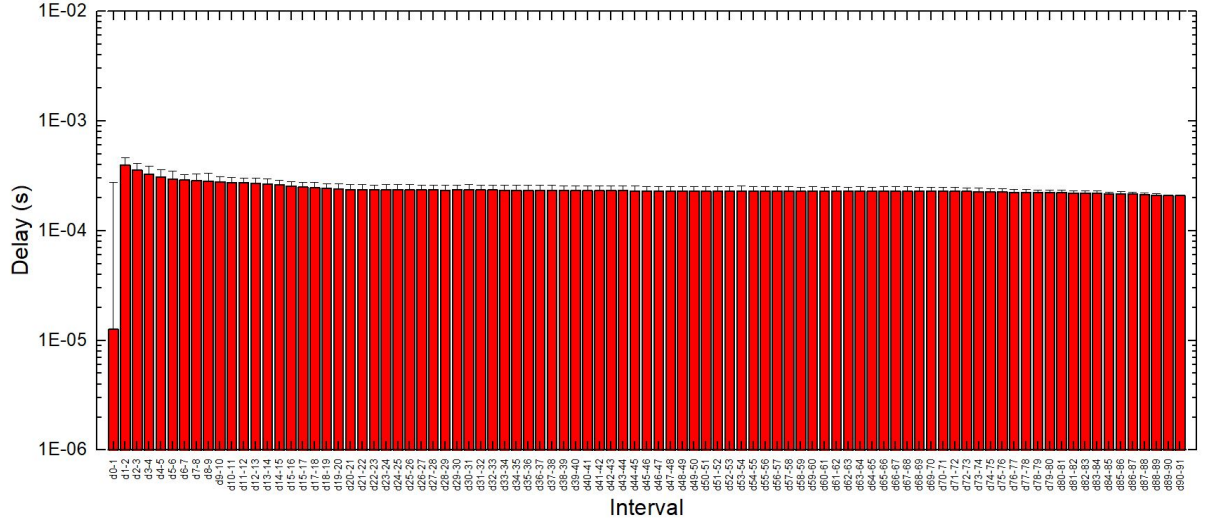


Figure A.3: Time interval between consecutive discharges during a single voltage pulse for positive streamers in ambient air. Measured with 22.5 kV at 900 mbar

A.1.2 Negative streamers in ambient air

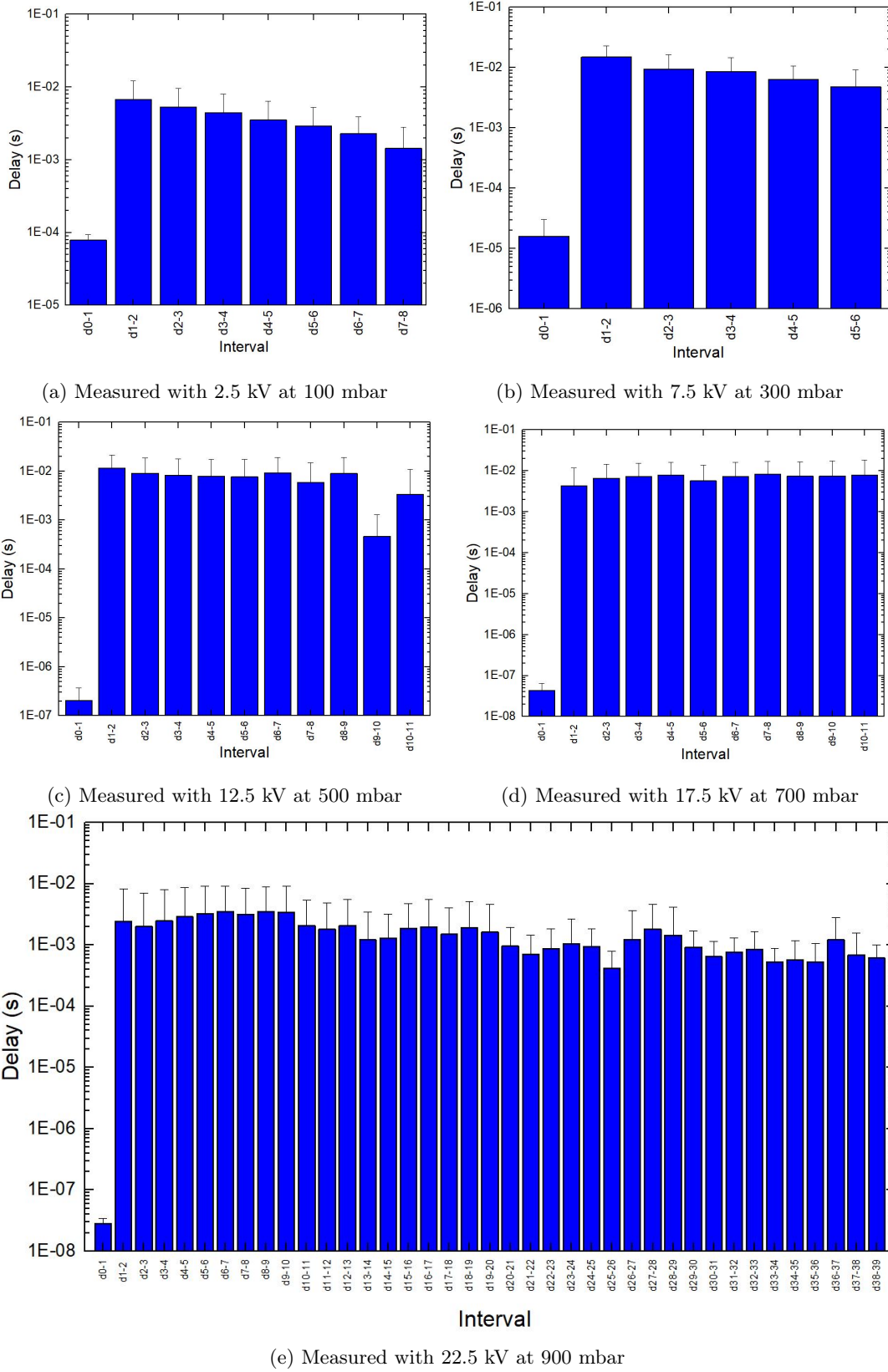
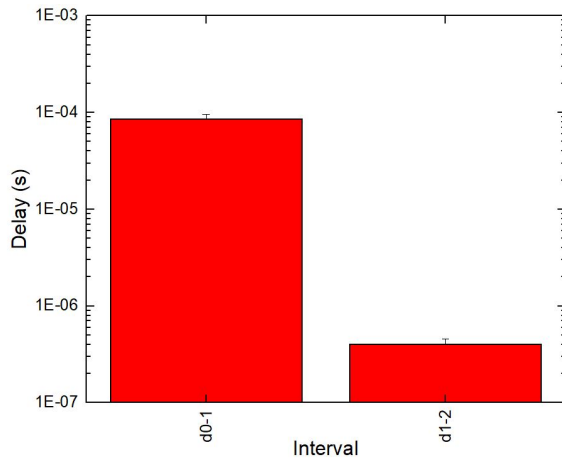


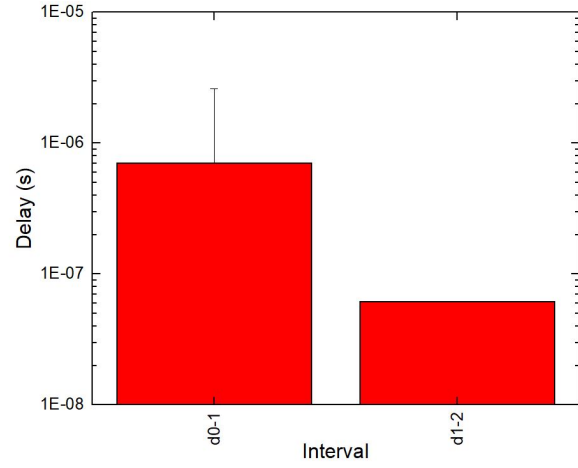
Figure A.4: Time interval between consecutive discharges during a single voltage pulse for negative streamers in ambient air.

A.1.3 Positive streamers in carbondioxide

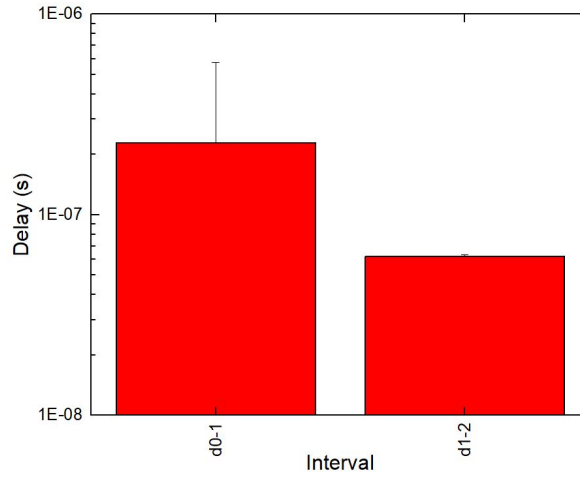
During these measurements the normal voltage values did not provide useful results, therefore other values were used.



(a) Measured with 7.7 kV at 300 mbar



(b) Measured with 17 kV at 700 mbar



(c) Measured with 21 kV at 900 mbar

Figure A.5: Time interval between consecutive discharges during a single voltage pulse for positive streamers in CO_2 .

A.1.4 Negative streamers in carbondioxide

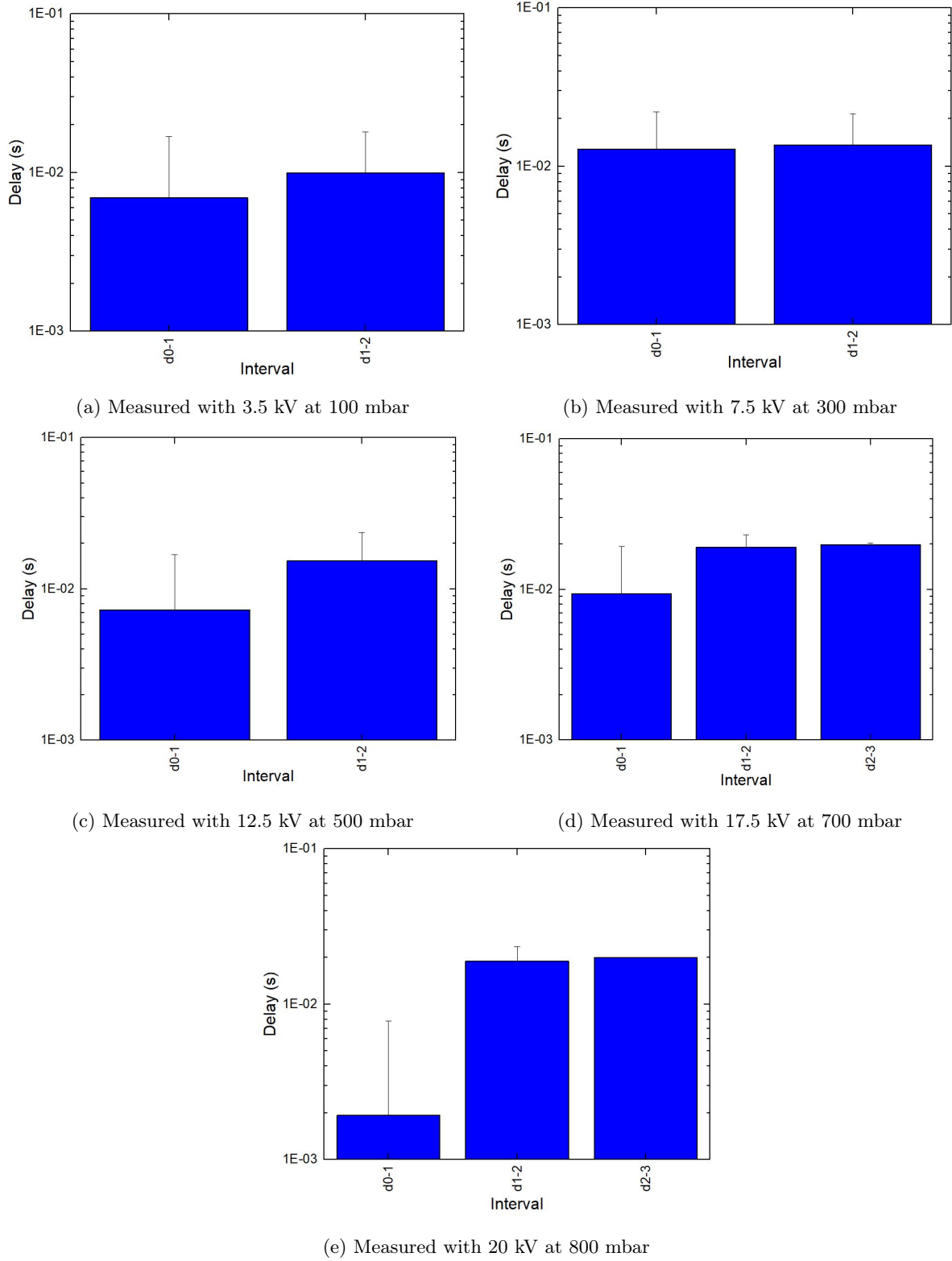
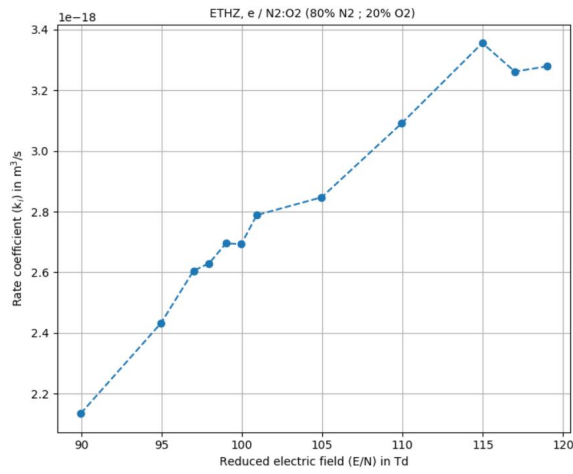


Figure A.6: Time interval between consecutive discharges during a single voltage pulse for positive streamers in CO_2 .

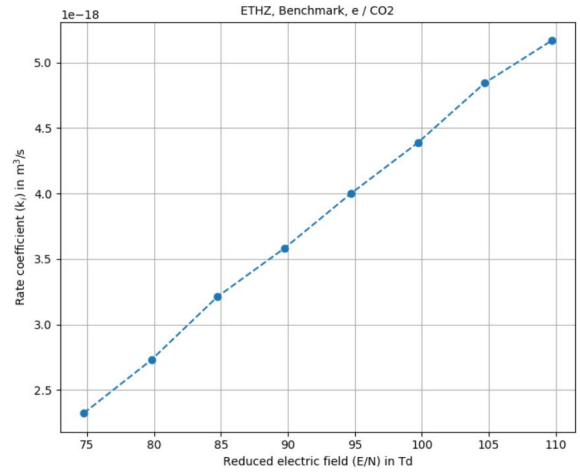
Appendix B

Swarm parameters

B.1 Electron attachment rate



(a) Air



(b) CO₂

Figure B.1: The electron attachment rate for different values of the reduced electric field. [5]

B.2 Recombination reaction rates

Table B.1: Recombination reaction rate coefficients for air and CO₂ ions and molecules. The unit is m³/s for two-body reactions and m⁶/s for three-body reactions.

Reaction	Rate coefficient	Reference
$\text{N}_2^+ + \text{e} \rightarrow \text{N} + \text{N}(^2\text{D})$	3.7×10^{-13}	[32]
$\text{N}_2^+ + \text{e} \rightarrow \text{N}_2$	1.5×10^{-13}	[32]
$\text{N}_2^+ + \text{O}^- \rightarrow \text{N}_2 + \text{O}$	7.8×10^{-12}	[32]
$\text{N}_2^+ + \text{O}_2^- \rightarrow \text{N}_2 + \text{O}_2$	7.5×10^{-12}	[32]
$\text{N}_2^+ + \text{O}_3^- \rightarrow \text{N}_2\text{O} + \text{O}_2$	7.8×10^{-12}	[32]
$\text{N}_2^+ + \text{NO}^- \rightarrow \text{N}_2 + \text{NO}$	7.8×10^{-12}	[32]
$\text{N}_4^+ + \text{O}_2^- \rightarrow 2\text{N}_2 + \text{O}_2$	7.8×10^{-12}	[32]
$\text{N}_4^+ + \text{NO}^- \rightarrow 2\text{N}_2 + \text{NO}$	7.8×10^{-12}	[32]
$\text{N}_4^+ + \text{NO}_2^- \rightarrow 2\text{N}_2 + \text{NO}_2$	7.8×10^{-12}	[32]
$\text{O}_2^+ + \text{e} \rightarrow \text{O} + \text{O}(^1\text{D})$	2.1×10^{-13}	[32]
$\text{O}_2^+ + \text{e} \rightarrow \text{O}$	1.5×10^{-13}	[32]
$\text{O}_2^+ + \text{O}^- \rightarrow \text{O}_2 + \text{O}$	7.5×10^{-12}	[32]
$\text{O}_2^+ + \text{O}_2^- \rightarrow 2\text{O}_2$	7.8×10^{-12}	[32]
$\text{O}_2^+ + \text{O}_3^- \rightarrow 2\text{O}_2 + \text{O}$	7.8×10^{-12}	[32]
$\text{O}^+ + \text{O}^- \rightarrow 2\text{O}$	7.6×10^{-12}	[32]
$\text{O}^+ + \text{O}_2^- \rightarrow \text{O}_2 + \text{O}$	7.8×10^{-12}	[32]
$\text{O}^+ + \text{O}_3^- \rightarrow 2\text{O}_2$	7.8×10^{-12}	[32]
$\text{O}_4^+ + \text{e} \rightarrow 2\text{O}_2$	2.1×10^{-12}	[32]
$\text{O}_4^+ + \text{O}^- \rightarrow \text{O}_2 + \text{O}_3$	7.8×10^{-12}	[32]
$\text{O}_4^+ + \text{O}_2^- \rightarrow 2\text{O}_2 + 2\text{O}$	2.0×10^{-12}	[32]
$\text{O}_4^+ + \text{O}_3^- \rightarrow 2\text{O}_2 + \text{O}_3$	7.8×10^{-12}	[32]
$\text{O}_2^+ + \text{NO}^- \rightarrow \text{NO} + \text{O}_2$	7.8×10^{-12}	[32]
$\text{O}_2^+ + \text{NO}_2^- \rightarrow \text{NO}_2 + \text{O}_2$	7.8×10^{-12}	[32]
$\text{O}_2^+ + \text{NO}_3^- \rightarrow \text{NO}_2 + \text{O}_2 + \text{O}$	7.5×10^{-12}	[32]
$\text{O}^+ + \text{NO}^- \rightarrow \text{NO} + \text{O}$	7.8×10^{-12}	[32]
$\text{O}^+ + \text{NO}_2^- \rightarrow \text{NO} + \text{O}_2$	7.8×10^{-12}	[32]
$\text{O}_4^+ + \text{NO}^- \rightarrow \text{O}_2 + \text{NO}_3$	7.8×10^{-13}	[32]
$\text{O}_4^+ + \text{NO}_2^- \rightarrow 2\text{O}_2 + \text{NO}_2$	7.8×10^{-13}	[32]
$\text{CO}_2^+ + \text{e} \rightarrow \text{CO}_2$	6.5×10^{-7}	[33]
$\text{CO}_2^+ + \text{e} \rightarrow \text{CO} + \text{O}$	6.5×10^{-7}	[33]
$\text{CO}_2^+ + \text{O}_2^- \rightarrow \text{CO} + \text{O}_2 + \text{O}$	6.0×10^{-7}	[33]

Appendix C

Electric field

C.1 Normal electric field in kV/cm

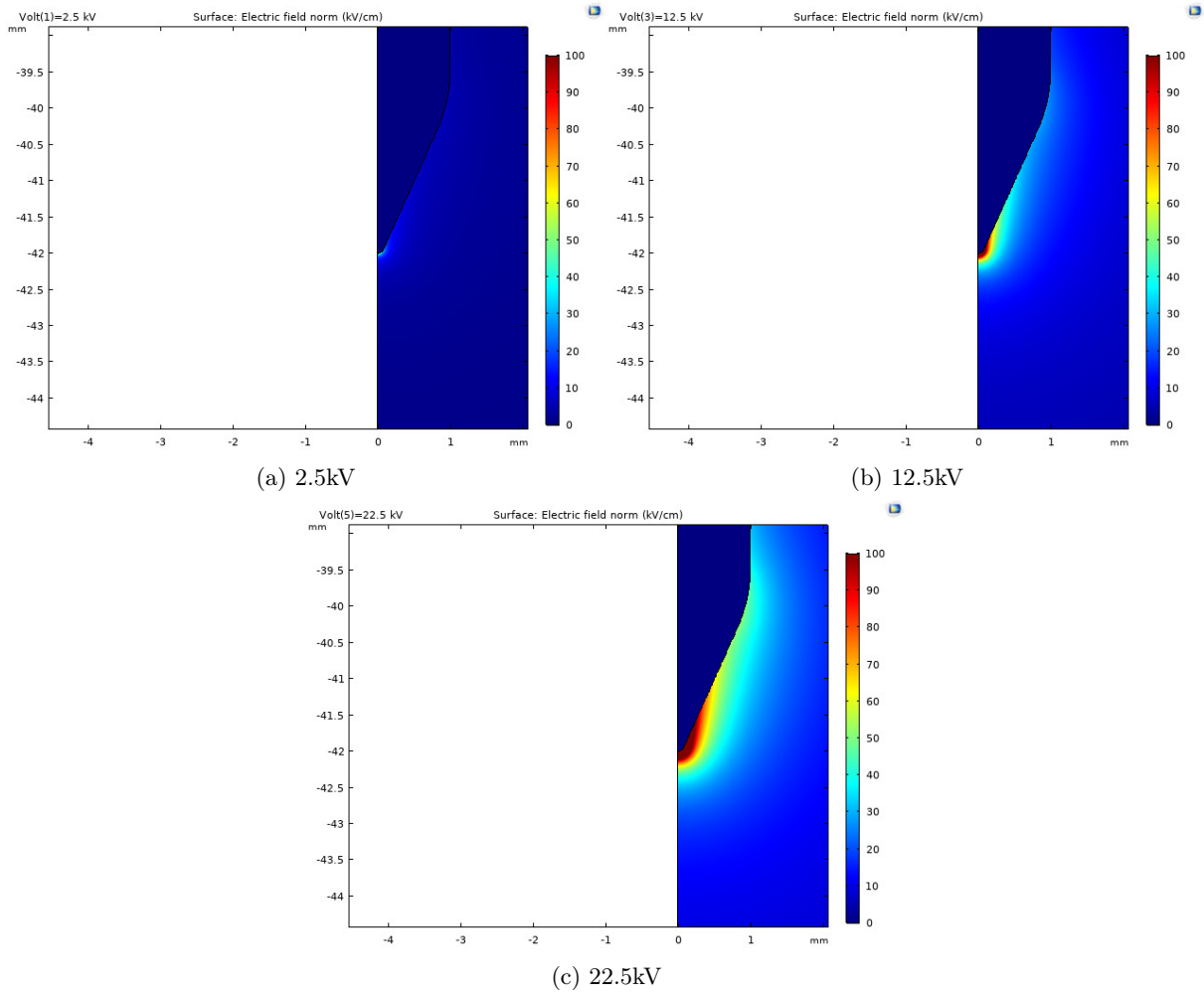


Figure C.1: The electric field in kV/cm for different values of the applied HV pulse.

C.2 Reduced electric field in Td

C.2.1 Air

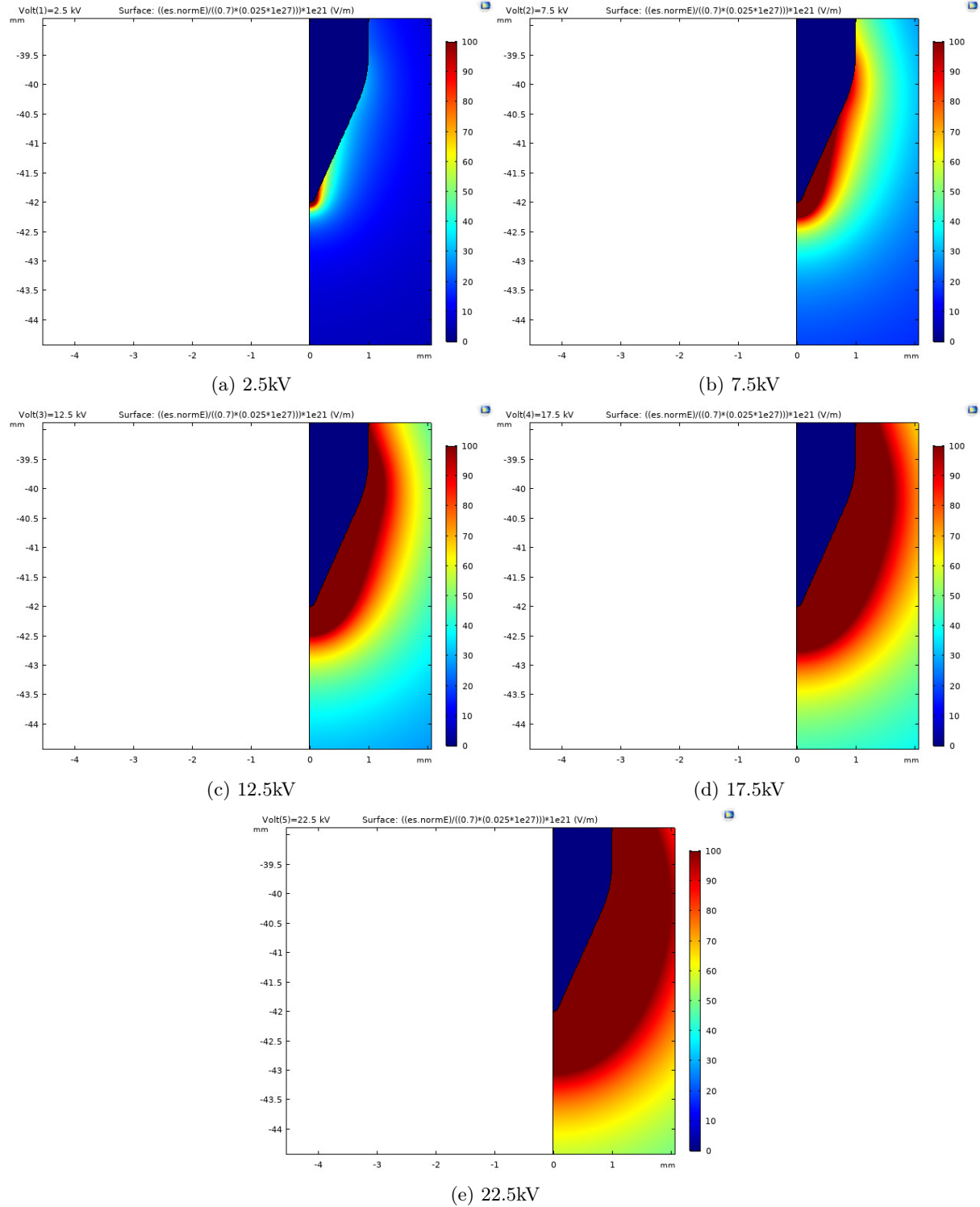


Figure C.2: The reduced electric field (unit Td) in air for different values of the applied HV pulse.

C.2.2 Carbondioxide

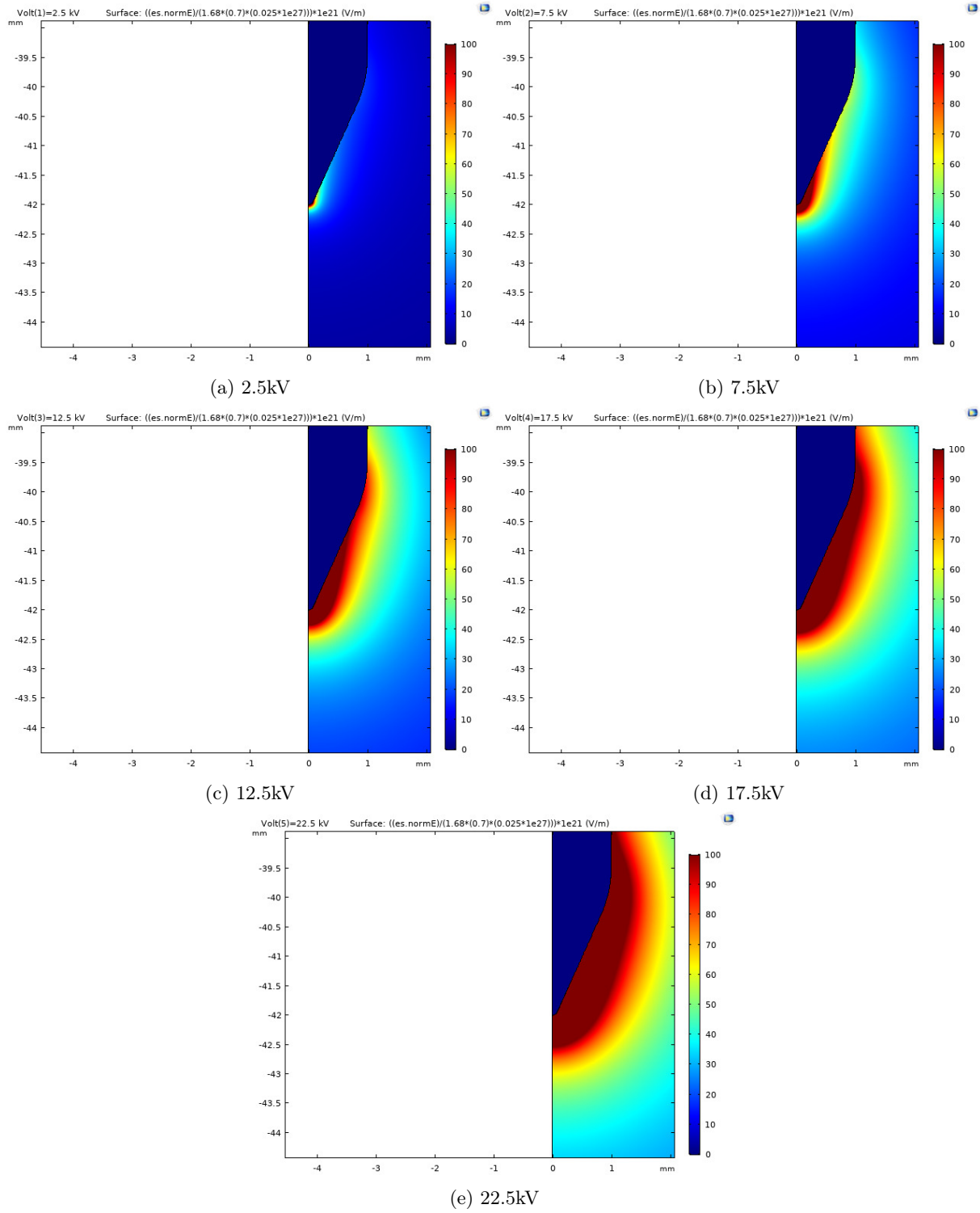


Figure C.3: The reduced electric field (unit Td) in CO₂ for different values of the applied HV pulse.