# 3D particle-in-cell simulations of negative and positive streamers in $C_4F_7N$ - $CO_2$ mixtures

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Abstract. We investigate negative and positive streamers in C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub> mixtures through simulations. These mixtures are considered to be more environmentally friendly than the insulating gas  $SF_6$  that is widely used in high voltage technology. Simulations are performed using a 3D particle-in-cell model. Negative streamers can propagate when the background field is close to the critical field. We relate this to their short conductive channels, due to rapid electron attachment, which limits their field enhancement. Positive streamers also require a background field close to the critical field, and in addition a source of free electrons ahead of them. In our simulations these electrons are provided through an artificial stochastic background ionization process as no efficient photoionization process is known for these gases. In 3D, we can only simulate the early inception stage of positive discharges, due to the extremely high electric fields and electron densities that occur. Qualitative 2D Cartesian simulations show that the growth of these discharges is highly irregular, resulting from incoming negative streamers that connect to existing channels. The inclusion of a stochastic background ionization process also has an interesting effect on negative discharges: new streamers can be generated behind previous ones, thereby forming a chain of negative streamers.

#### 1. Introduction

# 1.1. Eco-friendly alternative to the greenhouse gas $SF_6$

Sulfur hexafluoride  $(SF_6)$ , an insulating gas, has been widely used in electric power equipment such as gas circuit breakers and gas-insulated switchgear [1], due to its excellent properties for electrical insulation and current interruption [2,3]. But it is also the most potent industrial greenhouse gas, with a global warming potential (GWP) of about 23500 times that of  $CO_2$  over a 100-year horizon and an atmospheric lifetime of about 1000 years [4, 5]. Therefore there is an urgent need for exploring more sustainable alternatives. The most promising candidate is perfluoronitrile  $(C_4F_7N)$ , an electronegative gas developed by the 3M Company [6]. It has a relatively low GWP of about 1490, a relatively short atmospheric lifetime of about 22 years, good material compatibility with most electric power equipment, and a dielectric strength twice that of  $SF_6$  [7]. Depending on the application,  $C_4F_7N$ is often mixed with buffer gases such as  $CO_2$ ,  $N_2$  or dry air, primarily due to its relatively high liquefaction temperature (e.g. -4.7 °C at 0.1 MPa), considerations regarding environmental sustainability and safety, as well as cost reduction and availability [8].

# 1.2. Streamer discharges in $C_4F_7N$ - $CO_2$ mixtures

Numerous theoretical and experimental investigations have been conducted to explore the electric breakdown and recovery properties of  $C_4F_7N$  mixtures for practical applications. The decomposition pathways of  $C_4F_7N$  have been studied in [9–14]. Swarm parameters such as ionization and attachment coefficients have been experimentally measured to determine the reduced critical electric field and to estimate the dielectric strength of C<sub>4</sub>F<sub>7</sub>N mixtures [15–20]. Furthermore, their breakdown characteristics and dielectric properties have been extensively investigated through experimental measurements [21–30] as well as theoretical calculations [31-33]. C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub> mixtures with approximately 10%-20% C<sub>4</sub>F<sub>7</sub>N have been found to achieve a comparable dielectric strength to  $SF_6$  under varying conditions.

However, there are very few experimental studies on streamer discharges in  $C_4F_7N$ - $CO_2$  mixtures. This is primarily due to the challenge of imaging such discharges, given the lack of detectable light emission. Understanding streamer discharges in these mixtures is important, as they play a key role in the initial stage of electric breakdown [34]. Several authors have therefore computationally studied streamers in  $C_4F_7N$ - $CO_2$  mixtures with 2D fluid simulations [35–40], which will be further elaborated in section 5. The goal of this paper is to computationally study both negative and positive streamers in  $C_4F_7N$ - $CO_2$  mixtures in a full 3D geometry. In contrast to previous simulations in these mixtures, we use a 3D particle model which can capture the stochastic nature and branching of streamers in a more realistic way.

### 1.3. Cross sections for $C_4F_7N$

A set of electron-neutral collision cross sections is required as the model input for accurate modeling of streamers in  $C_4F_7N$ - $CO_2$  mixtures [41]. In recent years, numerous studies have been dedicated to obtaining cross section data for  $C_4F_7N$  through both theoretical and experimental methods [42].

The total electron impact ionization cross section of C<sub>4</sub>F<sub>7</sub>N was calculated using the Deutsch-Märk (DM) formalism [43] and the binary-encounter-Bethe (BEB) method [44]. To improve the agreement between these two methods. Zhong et al proposed to combine the DM and BEB formalisms with a dual sigmoid function [45]. In [46], the total and partial ionization cross sections of C<sub>4</sub>F<sub>7</sub>N were theoretically and experimentally studied. Furthermore, Chachereau et al estimated the total electron attachment cross section of C<sub>4</sub>F<sub>7</sub>N by inversely calculating swarm parameters obtained from a pulsed Townsend experiment [16]. The dissociative electron attachment process was also experimentally examined in [47], but no cross section data were given. In [48], Sinha et al utilized the spherical complex optical potential (SCOP) formalism to compute the total inelastic cross section of  $C_4F_7N$ , from which the electronic excitation and ionization cross sections were derived. The SCOP method was further used to calculate the elastic cross section of  $C_4F_7N$  in [49]. Note that the above-mentioned cross sections were all considered individually.

A complete set of electron-neutral collision cross sections for  $C_4F_7N$  has recently been proposed for the first time in [50], see figure 1, which will be used in the present paper. Beyond electron-neutral collisions, the ion kinetics including detachment and ion conversion may also influence the dielectric properties of  $C_4F_7N$ - $CO_2$  mixtures, but there is currently only limited data available on these processes [51].

### 2. Simulation method

We simulate negative and positive streamers in  $C_4F_7N-CO_2$  mixtures at 300 K and 1 bar. Simulations are performed with a 2D/3D PIC-MCC (particle-in-cell, Monte Carlo collision) model, using the open-source **Afivo-pic** code [52]. Below we give a brief summary of the model, see more information in [52, 53].

#### 2.1. PIC-MCC model

2.1.1. Particle mover and collisions Electrons are tracked as particles. Ions are included as densities and assumed to be immobile on the considered short time scales (up to 50 ns). Neutral gas molecules are present as a homogeneous background that electrons stochastically collide with.

In the electrostatic approximation, the positions  $\mathbf{x}$  and velocities  $\mathbf{v}$  of simulation particles are advanced in time with the 'Velocity Verlet' scheme [54] as

$$\mathbf{x}(t + \Delta t) = \mathbf{x}(t) + \Delta t \,\mathbf{v}(t) + \frac{1}{2} (\Delta t)^2 \,\mathbf{a}(t) \,, \tag{1}$$

$$\mathbf{v}(t + \Delta t) = \mathbf{v}(t) + \frac{1}{2}\Delta t \left[\mathbf{a}(t) + \mathbf{a}(t + \Delta t)\right], \qquad (2)$$

where  $\mathbf{a} = -(e/m_{\rm e}) \mathbf{E}$  is the acceleration due to the electric field  $\mathbf{E}$ , e the elementary charge, and  $m_{\rm e}$  is the electron mass.

Electron-neutral collisions are treated using the null-collision method [55] assuming isotropic scattering, with collision rates derived from cross sections, see section 2.2.1. We only consider electron-neutral collisions since the ionization degree of the discharges is typically below  $10^{-4}$ .

2.1.2. Super-particles In the PIC-MCC model, the stochastic development of discharges can be effectively captured by tracking individual electrons' trajectories and interactions. However, it is computationally infeasible to simulate every electron individually due to the large number of electrons (above  $10^8$ ) in a typical discharge. To address this, super-particles are used, with their weights  $w \ (w \ge 1)$  determining the number of physical electrons they represent. Such super-particles can be merged or split between time steps using adaptive particle management [56], thereby adjusting their weights w to a desired weight  $w_d$  as

$$w_{\rm d} = n_{\rm e} \,\Delta V / N_{\rm ppc} \,, \tag{3}$$

where  $n_{\rm e}$  is the electron density in a grid cell,  $\Delta V$  the cell volume, and  $N_{\rm ppc}$  is the desired number of particles per cell, which is here set to  $N_{\rm ppc} = 100$ .

Therefore, super-particles at low electron densities represent few (or even single) physical electrons, whereas those at high electron densities represent multiple physical electrons.

2.1.3. Adaptive mesh refinement for the electric field Simulation particles are mapped to grid densities using a standard bilinear (2D) or trilinear (3D) weighting scheme. Near refinement boundaries, the mapping is locally switched to the 'nearest grid point' scheme, to maintain the conservation of particle densities [53].



Figure 1: A complete set of electron-neutral collision cross sections for  $C_4F_7N$  from [50]. This set includes elastic cross section ( $Q_{elas}$ ), electron attachment cross sections ( $Q_{att1}$ ,  $Q_{att2}$ ), vibrational excitation cross sections ( $Q_{vib1}$ ,  $Q_{vib2}$ ), electronic excitation cross section ( $Q_{exc}$ ) and electron impact ionization cross section ( $Q_{ion}$ ).

Subsequently, the electric field **E** is calculated as  $\mathbf{E} = -\nabla \phi$ . The electric potential  $\phi$  is obtained by solving Poisson's equation

$$\nabla^2 \phi = -\rho/\varepsilon_0 \,, \tag{4}$$

where  $\rho$  is the space charge density and  $\varepsilon_0$  is the vacuum permittivity. Equation (4) is solved using the geometric multigrid method included in the Afivo library [57, 58]. The calculated electric field is then interpolated from the grid back to particles using standard bilinear or trilinear interpolation.

For computational efficiency, the Afivo-pic code includes adaptive mesh refinement. The mesh is refined if

$$\alpha_{\rm eff}(E)\Delta x > 1\,,$$

where  $\Delta x$  is the grid spacing and  $\alpha_{\text{eff}}(E) = \alpha(E) - \eta(E)$  is the field-dependent effective ionization coefficient, see figure 2. The minimal grid spacing is less than  $1 \,\mu\text{m}$  in the simulations. Note that we here use the effective ionization coefficient  $\alpha_{\text{eff}}(E)$  instead of the ionization coefficient  $\alpha(E)$  utilized previously in [52, 53].

#### 2.2. Input data

2.2.1. Cross sections We use cross sections for elastic, vibrational excitation, electronic excitation, ionization and attachment collisions for  $C_4F_7N$  from the XJTUAETLab database [50, 59], as shown in figure 1. This database provides a complete cross section set for  $C_4F_7N$ .

Electron-neutral collision cross sections for  $CO_2$ were obtained from the IST-Lisbon database [60].

Gas	Critical electric field $E_{\rm k}$
air	$28.2 \mathrm{kV/cm}$
$1\% C_4 F_7 N-99\% CO_2$	$40.2 \mathrm{kV/cm}$
$2\% C_4 F_7 N-98\% CO_2$	$47.2 \mathrm{kV/cm}$
$5\% C_4 F_7 N-95\% CO_2$ $10\% C_4 F_7 N-90\% CO_2$	$61.0\mathrm{kV/cm}$ $76.4\mathrm{kV/cm}$
$15\% C_4 F_7 N-85\% CO_2$	88.6 kV/cm
$20\% C_4 F_7 N-80\% CO_2$	99.7 kV/cm

Table 1: The critical electric field  $E_k$  for different gases at 300 K and 1 bar, corresponding to figure 2.

This database includes an effective momentum-transfer cross section, considering both elastic and inelastic processes [61]. For PIC simulations, an estimated elastic cross section was obtained by subtracting the total inelastic cross sections.

For comparison of electron transport data in different gases, electron-neutral collision cross sections for air were taken from the Phelps database [62].

2.2.2. Electron transport data Differences in electron transport data for various  $C_4F_7N$ - $CO_2$  mixtures, pure  $CO_2$  and air are illustrated in figure 2. These transport coefficients were computed with BOLSIG+ [63] at 300 K and 1 bar, using cross sections described in section 2.2.1. We remark that in the PIC-MCC model, these transport coefficients are not used since electron-neutral collisions are directly determined by cross sections.

Figure 2 shows that the ionization coefficient  $\alpha$ , flux mobility  $\mu$  and flux transverse diffusion coefficient  $D_{\rm T}$  are similar between these different gases. However, the attachment coefficients  $\eta$  in C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub> mixtures are several orders of magnitude higher than those in pure CO<sub>2</sub> and air, leading to higher critical electric fields  $E_{\rm k}$  (where the ionization coefficient  $\alpha$  is equal to the attachment coefficient  $\mu$ ), as summarized in table 1.

#### 2.3. 3D computational domain

We simulate negative and positive streamers in  $C_4F_7N-CO_2$  mixtures at 300 K and 1 bar using the 3D computational domain illustrated in figure 3(a), which measures  $5 \text{ mm} \times 5 \text{ mm} \times 10 \text{ mm}$ . The domain has a plate-plate geometry with a centrally positioned electrode protruding from the upper plate. The electrode is rod-shaped with a semi-spherical tip, measuring 2 mm in length and 0.4 mm in diameter.

For the electric potential, a constant high voltage V is applied on the upper plate and the rod electrode, the lower plate is grounded, and homogeneous

Neumann boundary conditions are applied on the other sides of the domain. Simulation particles are removed from the simulation if they enter the electrode or move beyond the domain boundaries. Secondary electron emission from the electrode is not included.

The axial electric field  $E_{\rm ax}(z)$  away from the rod electrode relaxes to the average field between the two plate electrodes, see figure 4. We therefore define the background electric field  $E_{\rm bg}$  as

$$E_{\rm bg} = |V|/d_{\rm plates} \,, \tag{5}$$

where  $d_{\text{plates}} = 10 \text{ mm}$  is the distance between the two plate electrodes. Note that  $E_{\text{ax}}(z) \approx E_{\text{bg}}$  in most of the domain. In addition, there is the average electric field  $E_{\text{avg}}$  between the needle tip and the grounded electrode (as used in e.g. [64, 65]):

$$E_{\rm avg} = |V|/d_{\rm gap} \,, \tag{6}$$

where  $d_{\text{gap}} = 8 \text{ mm}$ . In our simulations we therefore have  $E_{\text{avg}} = 1.25 E_{\text{bg}}$ .

Note that the computational domain is relatively narrow. Since we use Neumann boundary conditions for the electric potential, the discharge will develop as if identical discharges were developing around it, by mirroring the computational domain in the x and ydirections. This tends to somewhat artificially confine the discharge in the x and y directions. However, we opted for such a domain since it reduces the rather high computational costs of the simulations.

#### 2.4. 2D computational domain

As will be discussed in section 4.1, we are only able to simulate the early inception stage of positive discharges in 3D. To qualitatively illustrate how such positive discharges could develop, we perform 2D Cartesian simulations using the computational domain shown in figure 3(b). This domain measures  $(10 \text{ mm})^2$  and has the same electrode geometry and boundary conditions as the 3D domain discussed above.

#### 2.5. Included free electron sources

In the simulations, we consider two sources of (initial) free electrons: a plasma seed near the electrode tip or stochastic background ionization, which will be explained below. With a plasma seed, we are able to start negative discharges that continue to propagate, but not positive ones. Positive discharges require a source of free electrons ahead of them for their propagation, but there is considerable uncertainty in the mechanisms that could provide such free electrons in  $C_4F_7N-CO_2$  mixtures.

Two potential free electron sources are background ionization and photoionization. Due to the fast attachment of electrons to  $C_4F_7N$ , background ionization would be present in the form of positive and neg-



Figure 2: Comparison of electron transport data in air, pure  $CO_2$  and  $C_4F_7N$ - $CO_2$  mixtures. (a) Ionization coefficient  $\alpha$ , (b) attachment coefficient  $\eta$ , (c) flux mobility  $\mu$  and (d) flux transverse diffusion coefficient  $D_T$ . These coefficients were computed with BOLSIG+ [63] at 300 K and 1 bar, with cross sections for artificial dry air (80% N<sub>2</sub> and 20% O<sub>2</sub>) obtained from the Phelps database [62], cross sections for  $CO_2$  obtained from the IST-Lisbon database [60], and cross sections for  $C_4F_7N$  obtained from the XJTUAETLab database [59].

ative ions. Although there is evidence of electron detachment in high electric fields and low pressures in pure  $C_4F_7N$  [51], it is not yet known which ions would form in  $C_4F_7N$ -CO<sub>2</sub> mixtures at atmospheric pressure and how easily electrons would detach from these ions. Even less is known about photoionization in these mixtures. However, since photoionization in CO<sub>2</sub> is much weaker than in air, due to the lower production of ionizing photons and their significantly smaller absorption distance [66], we expect photoionization to be weak in  $C_4F_7N$ -CO<sub>2</sub> mixtures as well.

Due to the above uncertainties we decided to include a simple stochastic background ionization process in some of our simulations, to qualitatively illustrate how discharges would develop with a continuous free electron source. Electron-ion pairs are produced in the whole computational domain at a rate  $k_0$  between  $10^{18} \text{ m}^{-3} \text{s}^{-1}$  and  $10^{19} \text{ m}^{-3} \text{s}^{-1}$ . Note that these amounts correspond to  $1 \text{ mm}^{-3} \text{ns}^{-1}$ and  $10 \text{ mm}^{-3} \text{ns}^{-1}$ , respectively. This production rate is high compared to laboratory experiments with a radioactive admixture [67], but much lower than the production rate of photoelectrons in air-like mixtures (for reference, in atmospheric air the photoionization rate is typically between  $10^{22} \text{ m}^{-3} \text{s}^{-1}$  and  $10^{24} \text{ m}^{-3} \text{s}^{-1}$  one millimeter away from a streamer discharge).

In the rest of our simulations (without stochastic background ionization), we only include an initial neutral seed near the tip of the electrode. Electronion pairs are then generated according to a Gaussian distribution as

$$n_{\rm e}(\mathbf{r}) = n_{\rm i}^+(\mathbf{r}) = N_0 \exp\left[-\frac{(\mathbf{r} - \mathbf{r}_0)^2}{\sigma^2}\right],\qquad(7)$$

where  $N_0 = 2 \times 10^{11} \,\mathrm{m}^{-3}$  (unless specified otherwise),  $\sigma = 0.2 \,\mathrm{mm}$ , and  $\mathbf{r_0}$  is the position of the electrode tip located at  $z = 8 \,\mathrm{mm}$ . The expected number of electrons in such a seed is  $\pi^{3/2} \sigma^3 N_0$ , which is about 9 with the above values.



Figure 3: A view of (a) the 3D  $(5 \text{ mm} \times 5 \text{ mm} \times 10 \text{ mm})$  and (b) the 2D  $(10 \text{ mm})^2$  Cartesian computational domains. Panel (b) also shows the electric field profile E without a discharge at an applied voltage V = -36 kV. The centrally positioned rod electrode, protruding from the upper plate, has a length of 2 mm and a diameter of 0.4 mm. Boundary conditions for the electric potential  $\phi$  are indicated.



Figure 4: Axial electric field distribution  $E_{\rm ax}(z)$  without a discharge for an applied voltage V = -36 kV. The background electric field  $E_{\rm bg}$  and the average electric field  $E_{\rm avg}$  are also indicated.

#### 3. Negative streamers in $C_4F_7N-CO_2$ mixtures

#### 3.1. Streamers with an initial neutral seed

We first present 3D simulation results of negative streamers that start from an initial neutral seed placed near the electrode tip.

3.1.1. Effect of the background electric field Figure 5 illustrates the effect of the background electric field

 $E_{\rm bg}$  on negative streamers in a 1% C<sub>4</sub>F<sub>7</sub>N-99% CO<sub>2</sub> mixture. First, in figure 5(b) we look into the case of a background field of  $E_{\rm bg} = 36 \,\rm kV/cm$ , which corresponds to  $E_{\rm avg} = 45 \,\rm kV/cm$ . For comparison, the critical field  $E_{\rm k}$  is about 40 kV/cm. At  $t = 1 \,\rm ns$ , electrons provided by the initial neutral seed initiate a streamer near the electrode due to the locally high electric field. The negative discharge grows at an almost constant velocity of about  $0.5 \times 10^6 \,\rm m/s$  until it crosses the gap at  $t = 19 \,\rm ns$ . Multiple branches form during its propagation.

Electrons are primarily observed around the streamer head, where the maximal electron density is about  $10^{20}$  m<sup>-3</sup>, as those inside the streamer channel rapidly attach to C<sub>4</sub>F<sub>7</sub>N. Only a short part of the channel behind the streamer head therefore has a significant electron conductivity, with the electric field in the channel approximately relaxing back to the background electric field, as shown in figure 6. To better show the discharge trajectory and morphology, the positive ion density  $n_i^+$  is also included in figure 5 as well as subsequent figures. Note that the color scale in these visualizations can affect apparent width of the channels, as illustrated in Appendix A.

In figure 5(a), the background electric field is decreased to  $E_{\rm bg} = 34 \, \rm kV/cm$ , corresponding to  $E_{\rm avg} = 42.5 \, \rm kV/cm$ . The negative discharge now decelerates and fades out before reaching the lower plate, losing its field enhancement, similar to fading negative streamers in air [69]. This is initially surprising, since the average



Figure 5: Effect of the background electric field  $E_{\rm bg}$  on the propagation of negative streamers in a 1% C<sub>4</sub>F<sub>7</sub>N-99% CO<sub>2</sub> mixture with an initial neutral seed using 3D simulations. The upper and lower plate electrodes are here and afterward respectively indicated by a dashed line. Shown is the time evolution of (1) the electron density  $n_{\rm e}$  and (2) the positive ion density  $n_{\rm i}^+$  through Visit's [68] 3D volume rendering; the opacity is indicated in the legend. The same visualization is also applied to subsequent 3D simulations. A short conductive channel is observed behind the streamer head due to the fast attachment of electrons to C<sub>4</sub>F<sub>7</sub>N, so that the background field required to cross the gap is approximately the critical field  $E_{\rm k} \approx 40 \,\rm kV/cm$ . We remark that the maximal electron density is about  $10^{20} \,\rm m^{-3}$ , which is well above the limit of the color scale.

electric field  $E_{\rm avg} = 42.5 \, \rm kV/cm$  exceeds  $E_{\rm k}$ . However, this phenomenon can be explained by the fast decay of the electron conductivity in the channels, which leads to streamers with weak field enhancement and poorly conducting channels, as illustrated in figure 6. Streamers in strongly attaching gases thus modify the background electric field to a much smaller extent than streamers in gases such as air, which means that their propagation requires a background electric field close to the critical field  $E_{\rm k}$ . When  $E_{\rm avg} \approx E_{\rm k}$ , there is a region where the background field exceeds  $E_{\rm k}$  near the rod electrode but also a region below  $E_{\rm k}$  farther away from it, and streamers can stop propagating in the latter region, as shown in figures 5(a).

3.1.2. Effect of the initial neutral seed The streamers cross the gap in a background field  $E_{\rm bg} \approx 36 \, \rm kV/cm$ . This value is close to the critical field  $E_{\rm k}$ , as has also



Figure 6: Cross sections of the electric field E for the negative streamer shown in figure 5(b) at  $E_{\rm bg} = 36 \,\rm kV/cm$ . The electric field behind the streamer head approximately relaxes back to the background electric field due to rapid electron attachment. Note that the color scale ranges from 20 kV/cm to 100 kV/cm.

been observed in the electronegative gas  $SF_6$  [70]. The reason is that fast electron attachment shortens the conductive channel behind the streamer head, which strongly reduces the electric field enhancement at the head. (For comparison, the effect of strong attachment on axisymmetric streamers in air was studied in [71].)

From these results, one might conclude that the stability field  $E_{\rm st}$  is  $36 \, \rm kV/cm$ , if the stability field is defined as the background electric field required for a streamer to cross a gap [72]. However, we have shown recently in [69, 73] that the electric field where streamers in air propagate steadily, i.e. with constant shape and velocity, depends on their radius. So there is no unique stability field, but streamers with larger radii propagate steadily in lower background electric fields. A similar effect can be seen in figure 7. Here the same conditions are used as in figure 5(b), except that the maximal electron density  $N_0$  of the initial seed is changed from  $2 \times 10^{11} \,\mathrm{m^{-3}}$  to  $1 \times 10^{11} \,\mathrm{m^{-3}}$  and  $5 \times 10^{11} \,\mathrm{m^{-3}}$ , respectively. Although most discharges are able to cross the gap, we find that the larger seed creates a wider discharge containing more streamer channels, making it easier for the discharge to cross the gap in the same background field of  $E_{\rm bg}$  =  $36 \,\mathrm{kV/cm}$ . The average streamer velocity is also about 40% higher. This is due to a collective effect where neighboring branches mutually increase the electric field enhancement at the streamer head.

3.1.3. Effect of the  $C_4F_7N$  concentration The effect of the  $C_4F_7N$  concentration is illustrated in figure 8, where the C<sub>4</sub>F<sub>7</sub>N fraction is increased to 5%, 10% and 20%. These mixtures are of interest in practical applications as their dielectric strength is comparable to SF<sub>6</sub>. Given the sensitivity of negative streamer propagation to the background electric field  $E_{\rm bg}$ , here we set  $E_{\rm bg} = 0.9 E_{\rm k}$  for all cases, see table 1.

All negative streamers decelerate and fade out before crossing the gap, in contrast to the case with 1% C<sub>4</sub>F<sub>7</sub>N shown in figure 5(b), which is also at  $E_{\rm bg} = 0.9 E_{\rm k}$ . Furthermore, the streamer channels are thinner, slower and they branch more when the C<sub>4</sub>F<sub>7</sub>N fraction is increased, and they stop earlier. This indicates that for increased C<sub>4</sub>F<sub>7</sub>N concentrations the background electric field  $E_{\rm bg}$  required to cross the gap is above  $0.9 E_{\rm k}$ , due to a higher electron attachment rate that further decreases the streamer's electric field enhancement.

#### 3.2. Streamers with stochastic background ionization

We now perform 3D simulations of negative streamers in which the initial neutral seed is replaced by stochastic background ionization that continuously produces electron-ion pairs within the computational domain, as discussed in section 2.5. Although such background ionization is not required for negative streamer propagation, we do observe some interesting effects in figure 9, in which the background ionization rate  $k_0$  is varied. With a higher  $k_0$  more electronion pairs are produced per unit of time. As expected, this results in earlier initiation and the formation of a greater number of streamers.

A remarkable phenomenon is that new negative streamers are generated behind the previous ones. This happens because the electric field behind previous streamers quickly relaxes back to the background electric field, due to rapid electron attachment, which allows new streamers to form. This process repeats itself, at least within the time scales considered, so that a chain of negative streamers emerges. In some cases, later streamers are faster and they can approach or overtake previous ones, as shown in figure 9(a)for the streamers numbered (2) and (3). Although there is only a low electron conductivity behind the repeated streamers, the ion density and hence the ion conductivity increase over time, if the recombination of positive and negative ions is not too fast. If the ion conductivity keeps increasing, we expect that the ions eventually will screen the electric field and inhibit the formation of new streamers.

## 4. Positive streamers in $C_4F_7N-CO_2$ mixtures

In this section, we perform both 3D and 2D simulations of positive streamers in  $C_4F_7N$ - $CO_2$  mixtures. The simulations include stochastic background ionization,



Figure 7: Effect of the maximal electron density  $N_0$  of the initial seed on the propagation of negative streamers. All parameters are the same as in figure 5(b), except for  $N_0$ , which is changed from  $2 \times 10^{11} \text{ m}^{-3}$  to  $1 \times 10^{11} \text{ m}^{-3}$  and  $5 \times 10^{11} \text{ m}^{-3}$ , respectively. For both cases, four simulation runs with different numbers of initial electrons are shown. An increase of the density  $N_0$  leads to the formation of a wider and faster discharge containing more streamer channels, making it easier for the discharge to cross the gap. This resembles the observation that in air streamers with larger radii propagate steadily in lower background fields [69, 73].

which serves as the source of free electrons for positive streamer propagation, as discussed in section 2.5.

# 4.1. 3D simulation results

In 3D, we can only simulate the early inception stage of positive discharges. Two examples for  $k_0 = 1 \times 10^{19} \,\mathrm{m^{-3}s^{-1}}$  and  $k_0 = 5 \times 10^{19} \,\mathrm{m^{-3}s^{-1}}$  are shown in



Figure 8: Effect of the  $C_4F_7N$  concentration on the propagation of negative streamers at  $E_{bg} = 0.9 E_k$  with an initial neutral seed, see table 1. The maximal electron density is about  $10^{21} \text{ m}^{-3}$ . As the  $C_4F_7N$  concentration increases, the streamer branches into more channels, which are thinner, slower, and stop earlier.

figure 10. In these simulations, electron avalanches develop towards the electrode, which quickly transition into negative streamers. Since these negative streamers are much thinner than the electrode itself, the maximal electric field at the streamer heads becomes extremely high, exceeding  $10^8 \text{ V/m}$ , as illustrated in figure 11. This also results in very high electron densities, exceeding  $10^{23} \text{ m}^{-3}$ .

Computationally, it is very demanding to simulate discharges with such high fields and electron densities, due to the high grid resolution (below  $1 \,\mu$ m) and small time steps (below  $10^{-14}$  s) required. Although we are only able to simulate the first stages of inception, our results suggest that positive discharges develop in a

highly irregular way in  $C_4F_7N$ -CO<sub>2</sub> mixtures, with sharp features and very high fields at their tips. We speculate that the growth of positive discharges will be the result of incoming negative streamers that connect to an existing discharge channel. To qualitatively study such growth, we will perform 2D Cartesian simulations in section 4.2.

#### 4.2. 2D Cartesian simulation results

To qualitatively study the growth of positive discharges in  $C_4F_7N$ - $CO_2$  mixtures, we now perform 2D Cartesian simulations. Computational costs are much lower in such a geometry, not only because there is one



Figure 9: Effect of the background ionization rate  $k_0$  on the propagation of negative streamers without an initial neutral seed. The maximal electron density is about  $10^{20} \text{ m}^{-3}$ . With stochastic background ionization, a remarkable phenomenon occurs where new negative streamers are generated behind the previous ones, due to rapid electron attachment. This process repeats itself leading to the formation of a chain of negative streamers, each represented by a red dashed curve and sequentially numbered to indicate their order of emergence.

less dimension but also because field enhancement is significantly weaker, as there is no curvature in the third dimension. Although there are quantitative differences between 2D and 3D in terms of streamer properties and branching, 2D simulations can help to qualitatively understand the growth of positive discharges.

The interpretation of particle weights and stochastic fluctuations is somewhat complicated in 2D. We here use a minimum particle weight of  $w_{\min} = 10^5 \text{ m}^{-1}$ . This can for example be interpreted as having an unresolved "third" dimension in the simulations of 1 m and a minimum particle weight of  $10^5$ , or equivalently as having and a third dimension of 10  $\mu$ m and a minimum weight of one, see [74] for details. We will express the background ionization rate as  $k_0/w_{\rm min}$ , with units m<sup>-2</sup>s<sup>-1</sup>.

Figure 12 shows the time evolution of a positive streamer in a 1% C<sub>4</sub>F<sub>7</sub>N-99% CO<sub>2</sub> mixture with  $E_{\rm bg} = 36 \,\rm kV/cm$  ( $E_{\rm avg} = 45 \,\rm kV/cm$ ) and  $k_0/w_{\rm min} = 1 \times 10^{14} \,\rm m^{-2} s^{-1}$ . Electron avalanches form due to stochastic background ionization, and they initially develop towards the electrode from various directions. During their development, these avalanches transition into short negative streamers, which connect to each other, thereby extending the positive streamer channel. This process repeats over time, leading to the irregular and branched downward propagation of the positive



Figure 10: Two examples of positive discharges in 3D simulations using stochastic background ionization, without an initial neutral seed. Both the side and bottom views are presented, with a zoomed-in perspective centered at the electrode tip. Here we only show the time evolution of the electron density  $n_e$  in time steps of 0.025 ns through Visit's 3D volume rendering, with a linear scale ranging from 0 to  $1 \times 10^{21} \text{ m}^{-3}$ . The maximal electron density is about  $10^{24} \text{ m}^{-3}$ . In 3D, we are only able to simulate the early inception stage of positive discharges due to the presence of extremely high electric fields and electron densities.



Figure 11: Cross sections of (a) the electric field E and (b) the electron density  $n_{\rm e}$  for the two positive discharges shown in figure 10 at the last frame, with a  $(1 \text{ mm})^2$  zoomed-in perspective centered at the electrode tip. Note that  $n_{\rm e}$  is shown on a logarithmic scale.  $E_{\rm max}$  exceeds  $10^8 \text{ V/m}$ , and the maximal electron density exceeds  $10^{23} \text{ m}^{-3}$ .

streamer.

In figure 13 we present eight different positive streamers by varying the background electric field  $E_{\rm bg}$ , background ionization rate  $k_0/w_{\rm min}$  and  $C_4F_7N$ concentration. Results are shown at t = 50 ns, and the streamers in panels (d)–(h) have crossed the gap. In all cases, the positive channels extend by incoming negative streamers, as was illustrated by the temporal evolution in figure 12. Note that the number of streamer channels increases with the background field and with the background ionization rate. In 3D, we expect positive streamers to grow in a similar manner, with the channels extending by incoming negative streamers. However, the electric fields at the streamer tips would be much higher, and so would be the electron densities. Furthermore, one would generally expect more branching in 3D, although the number of branches would to some extent depend on the amount of background ionization. Such highly stochastic growth has been experimentally observed in gases with weak or no photoionization [75].



Figure 12: An example of a positive streamer using 2D Cartesian simulations with a background ionization rate  $k_0/w_{\rm min} = 1 \times 10^{14} \,\mathrm{m^{-2} s^{-1}}$ , without an initial neutral seed. Shown is the time evolution of (a) the electric field E, (b) the electron density  $n_{\rm e}$ , (c) the positive ion density  $n_{\rm i}^+$  and (d) the space charge density  $\rho$ . The growth of the positive discharge exhibits a highly irregular and branched structure, resulting from incoming negative streamers that connect to existing channels.



Figure 13: Positive ion density  $n_i^+$  for eight different positive streamers at t = 50 ns using 2D Cartesian simulations with stochastic background ionization, without an initial neutral seed. The background electric field  $E_{\rm bg}$ , background ionization rate  $k_0/w_{\rm min}$  and C<sub>4</sub>F<sub>7</sub>N concentration are varied. All the positive streamers develop in a highly irregular way, with the positive channels extending by incoming negative streamers.

#### 5. Comparison with past computational work

Below, we briefly discuss related computational work. To the best of our knowledge, all previous simulations of discharges in  $C_4F_7N$  mixtures were performed using 2D fluid models. In [36], streamers in  $C_4F_7N$  mixtures with different buffer gases were simulated at 300 K and 1 bar using an axisymmetric model [76]. Both ionization and attachment processes were considered, with their coefficients interpolated from [16, 19, 77]. Photoionization was not considered, but instead an initial homogeneous density of  $10^{14} \text{ m}^{-3}$  for electrons and positive ions was included. The results showed that a higher background field was required for streamers to propagate in  $C_4F_7N$ -N<sub>2</sub> than in  $C_4F_7N$ -CO<sub>2</sub>.

In [38], negative corona discharges in  $C_4F_7N-N_2$ and  $C_4F_7N-CO_2$  mixtures were simulated at 300 K and 4 bar using an axisymmetric model. The ionization and attachment cross sections of  $C_4F_7N$  were obtained from [45] and [16], respectively. Electron-ion and ion-ion recombination processes were also considered. The negative corona discharges started from an initial Gaussian seed, and no other background ionization was included. However, most of the negative discharges in the simulations remained in the inception stage and were far from propagation.

In [39], positive streamers in  $C_4F_7N$  were simulated in 2D with varying electrode shapes, using the same cross section data as [49]. An unspecified background ionization density was assumed as a replacement for photoionization. Increasing the applied voltage led to higher maximum electric fields and velocities. In another study [40], the authors performed 2D simulations of surface discharges in a 9%  $C_4F_7N-91\%$  CO<sub>2</sub> mixture at 300 K and 1 bar, again using an unspecified background ionization density. They investigated the effect of the applied voltage and the dielectric constant on the discharge, and compared surface discharges in  $C_4F_7N-CO_2$  with those in  $SF_6$ .

The main novelty of our work is that we for the first time use 3D particle simulations, with which we can capture the stochastic inception and branching of streamers in  $C_4F_7N$ -CO<sub>2</sub> mixtures. These stochastic aspects are particularly important since these mixtures appear to lack an effective photoionization mechanism, which results in much more irregular discharge growth than for example in air.

Another difference is that we have included a stochastic background ionization process in some of the simulations, which produces electron-ion pairs at a certain rate over time. In previous simulations of positive discharges, an initial background ionization density was assumed. We observed that an initial electron density would quickly decay due to rapid attachment (unless the background field  $E_{\rm bg}$  was above the critical field  $E_{\rm k}$ ), after which positive streamer propagation was hardly possible. However, both approaches are rather artificial, and further work is necessary to better understand free electron sources in such mixtures, such as photoionization and electron detachment from negative ions.

Our observation that streamers in C<sub>4</sub>F<sub>7</sub>N-CO<sub>2</sub> mixtures can only keep propagating if the background electric field  $E_{bg}$  is approximately the critical electric field  $E_{\mathbf{k}}$  (due to rapid electron attachment) is in agreement with previous work. However, the observed discharge growth in our 3D simulations differs significantly from that observed in previous 2D fluid simulations. First, we observe stochastic growth with frequent branching, both for negative and positive polarities. Second, we observe that new negative streamers can form behind previous ones, forming a chain of negative streamers. Third, in 2D Cartesian simulations we see that positive discharges extend due to incoming negative streamers that connect to existing channels.

#### 6. Conclusions and outlook

We have simulated negative and positive streamers in  $C_4F_7N$ - $CO_2$  mixtures at 300 K and 1 bar, using a 3D and 2D particle-in-cell model. Negative streamers were able to start from a small number of initial electrons near an electrode, whereas positive streamers required a sustained source of free electrons ahead of them for their propagation. We included an artificial stochastic background ionization process to provide such free electrons.

For negative streamers, a short conductive channel was observed behind the streamer head due to the fast attachment of electrons to  $C_4F_7N$ . Due to the short channel, these discharges did not gain as much field enhancement as streamers in most other gases, so the background field required for streamer propagation was approximately the critical electric field. Surprisingly, new negative streamers could be generated behind the previous ones when a stochastic background ionization process was included. In this manner a chain of negative streamers could form.

Simulating positive streamers required the inclusion of the artificial background ionization process, and it was much more challenging due to the presence of extremely high fields and electron densities. In 3D, we were only able to simulate the early inception stage of positive discharges. To qualitatively investigate the growth of these discharges, we performed 2D Cartesian simulations. We observed that positive discharge growth resulted from incoming negative streamers that connected to existing channels. This led to highly irregular discharge growth, in which branching was determined by the locations of free electrons ahead of the discharge.

If there is indeed no effective photoionization mechanism in  $C_4F_7N$ - $CO_2$  mixtures, our results suggest that negative streamers will propagate more easily than positive ones. This is in contrast to the behavior in air, where positive streamers initiate and propagate more easily than negative streamers [78,79].

**Outlook.** A better understanding of free electron sources such as photoionization and electron detachment is important for the modeling of streamer-like discharges in  $C_4F_7N$ - $CO_2$  mixtures. Including ion kinetics could also be important, as was demonstrated in [51]. It would be valuable if streamer propagation could be captured experimentally, so that simulations and experiments could directly be compared. Furthermore, a novel type of simulation model might be required to study positive discharges in these mixtures, because 3D simulations are currently too expensive due to the complex discharge structure and the extremely high electric fields and electron densities.

Finally, it would be interesting to consider additional ionization mechanisms. First, future research could include secondary electron emission due to ions to more realistically study the inception of negative streamers, as in e.g. [80]. Second, when the fraction of  $C_4F_7N$  or the pressure is increased together with the applied electric field, it could be interesting to include field ionization as in e.g. [81].

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#### Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: https://doi.org/10.5281/zenodo.8214598.

# Appendix A. Effect of the color scale on streamer visualization

Figure A1 presents results with different color scales for negative streamers shown in figure 5(b) at t = 10 ns, with the maximum scale value being  $1 \times 10^{19} \text{ m}^{-3}$ ,  $5 \times 10^{19} \text{ m}^{-3}$  and  $1 \times 10^{20} \text{ m}^{-3}$ , respectively. With a higher value of the maximum, the streamers appear to be thinner.

Throughout the paper, a linear scale ranging from 0 to  $1 \times 10^{19} \,\mathrm{m^{-3}}$  is used for the electron density  $n_{\rm e}$  and a linear scale ranging from 0 to  $1 \times 10^{20} \,\mathrm{m^{-3}}$  is used for the positive ion density  $n_{\rm i}^+$ , except for the 3D simulations of positive discharges shown in section 4.1, which have much higher densities, see figures 10 and 11.

# Appendix B. Computational costs

The primary drawback of a 3D PIC-MCC model is its high computational cost, due to the large number of simulation particles required. The simulations in this paper were performed on workstations with 8 cores and 32–64 GB of RAM, using at most  $5 \times 10^8$  simulation particles. The run time for the negative cases shown in section 3 was about 1 week, whereas the 3D simulations of positive streamer inception shown in section 4.1 took about 2 weeks. The 2D simulations of positive streamers shown in section 4.2 only took a few hours.

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Figure A1: The case of negative streamers in figure 5(b) at t = 10 ns. Shown is the electron density  $n_e$ , negative ion density  $n_i^-$  and positive ion density  $n_i^+$  through Visit's 3D volume rendering with different color scales. With a higher maximum value of the color scale streamer channels appear to be thinner.

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