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SIMULATION OF ELECTRON SWARM PARAMETERS IN SF_6

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The motion of electrons in sulfur hexafluoride in uniform electric fields is simulated using a Monte Carlo method. The evaluated swarm parameters are compared with experimental results for drift velocity, electron mean energy, ratio of ionization coefficient and attachment coefficient. The electron−molecule collision cross sections adopted in the simulation result are in a good agreement with the experimental values over the range of E/N investigated (E is the electric field and N is the gas number density of background gas molecules).

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1. Introduction

The continuous growth of applications of low temperature plasmas for semiconductor fabrication processes, high voltage devices and ion sources and ion mobility mass spectrometry [1−3], to name only a few, is accompanied by research on the fundamental processes that occur in these gas discharges. Thus, the improvements of the measurement and calculation of basic data such as cross sections, swarm and transport coefficients are essential for gaining a better understanding and modelling of these phenomena. For obvious engineering purposes, a study of the breakdown strength of gases is important and involves the electron drift velocity, the main energy and the ionization and attachment coefficients. These quantities are referred to generally as the electron swarm parameters [4−6].

To attain better results in these application fields, a quantitative understanding of the glow discharge is required. Especially interesting are the properties of discharges in electronegative gases, which are most frequently used for technological applications. Because of its outstanding electrical and physical properties, sulfur hexafluoride (SF_6) has been widely used in the electric power industry as an insulation medium for high voltage equipment. SF_6 presents excellent arc quenching properties, a breakdown strength three times higher than air at atmospheric pressure. It is a very stable electronegative (electron attaching), non toxic, and non flammable gas, and in its normal state, SF_6 is chemically inert. Its relatively large cross section for attaching low energy electrons under electrical stress inhibits the initiation and growth of electrical discharges. Moreover, due to the high etching rate of silicon in SF_6 plasma, the fabrication of integrated circuits is another recent use of SF_6 [7 − 10]. Since it has an excellent insulating property, SF_6 gas has contributed considerably to advances in miniaturization, and increasing high effectiveness and reliability of electric transmission and distribution instruments such as insulated switch gear (GIS).

Typically, electron swarm parameters are used in fluid simulations of plasmas to determine the time-dependent evolution of densities of radicals and charged particles; in particular, they are needed to solve the continuity equation for electrons, which includes drift, diffusion and electron multiplication processes [11]. Proper understanding of the electron dynamics in the plasma is essential since it is electron collisional processes that produce the radicals that act as etching or deposition precursors [12]. The numerical solution of the Boltzmann equation yields the electron energy distribution with the electric field E and gas number density N as parameters. Appropriate integration of the energy distribution function yields the transport and ionizing properties of the electron swarm. Monte Carlo simulation of electron drift in a uniform electric field has the advantage that the motion of the electron at all stages during its passage in the discharge is traced. In addition, the Monte Carlo method is easier to develop in hydrodynamic as well as non-hydrodynamic regimes.

The aim of this research is part of a long term project dealing with the determination of the electron swarm and transport parameters of the pure gas. In this paper, we have studied the behavior of electrons in uniform electric fields by a Monte Carlo method. Swarm parameters are determined as a function of E/N for different rates of increase of the electric field. The calculation has been performed for sulfur hexafluoride.

2. *Simulation method*

The electron transport in a gas under the influence of an electric field E can be simulated with the help of a Monte Carlo method [13 − 15]. Every electron, during its transit in the gas, performs a succession of free flights punctuated by elastic or inelastic collisions with molecules of gas defined by collision cross sections. During the successive collisions for every electron, certain information (velocity, position, etc.) is stored in order to calculate, from appropriate sampling methods, transport coefficients and macroscopic coefficients. In a spherical coordinate system, a background gas of SF_6 molecules with a number density of $N = 3.29 \times 10^{22}$ m^{-3}, which corresponds to a gas pressure of 133 Pa at 20°C, is considered. To avoid large negative powers of 10, a unit of 1 Td $= 10^{-21}$ Vm^2 is used. The applied electric field E is antiparallel to the z axis. n_0 electrons with a constant energy ε_0 are injected from the origin of the coordinate system assuming a cosine distribution for the angle of entry with respect to the z axis. At $t = 0$, an electron observes a free flight time with a randomly selected angle of entry, depending on the distribution. Interactions with the electrodes are not considered, and the collision is simulated by comparing the probability with computer generated random numbers at the end of each step. The "null collision" technique has been used, since the null collision method is computationally efficient. The null collision method involves the introduction of fictitious collisions in which no exchange of momentum or energy occurs, with cross sections chosen so that the total collision frequency becomes independent of velocity. In the Monte Carlo method, the electron moves in a time step dt or a distance ds under the force of the electric field E. The former approach is known as the flight time approach and the latter the free path approach. The above two methods have the disadvantage that the computational time required to calculate the motion of electrons is excessively long. This problem is overcome by the null collision technique. If we can find an upper bound of collision frequency $\nu_{\max}$ such that

$$\nu_{\max} = \max\{N, Q_{\rm t}(\varepsilon), W\}\,, \tag{1}$$

in which W is the drift velocity of an electron, $Q_{\rm t}$ is the total collision cross section in units of m^2, N is the gas number density and ε is the electron energy. The mean flight time is $1/\nu_{\max}$, and the time of flight

$$\mathrm{d}t = -\frac{\ln R_1}{\nu_{\max}}\,, \tag{2}$$

where R_1 is the random number uniformly distributed between 0 and 1. The assumed total collision cross section $Q'_{\rm t}$ (effective total cross section after introduction of "null process") is defined as

$$Q'_{\rm t} = Q_{\rm t} + Q_{\rm null}\,, \tag{3}$$

where $Q_{\rm null}$ is called the "null collision" cross section, and is the cross section for a fictitious process (null process) which causes no change in the properties of the electron. The new position and energy of the electron are calculated according to the equation of motion, assuming that the scattering is isotropic. If a collision is not observed, the direction is adjusted according to the parabolic orbit of the electron. The type of collision is simulated by comparison with the computer generated random numbers.

We can determine whether the collision is null or real after having determined that a collision takes place after a certain time interval dt. Let P_1 be the probability of a collision,

$$P_1 = \frac{Q_{\rm t}}{Q'_{\rm t}}. \tag{4}$$

For $P_1 > R_2$, where R_2 is a random number uniformly distributed between 0 and 1, the collision is real, otherwise the collision does not occur, and we proceed to the next collision without any change in electron energy and direction. For real collisions, the nature of the collision is determined in the following way: $P_{2,j}$ is the probability that collision process j takes place ($P_{2,1}$ is the probability for the null process), $j = 1, 2, 3, \dots, n$, including elastic, vibration, excitation, attachment and ionization collisions. The jth collision process is defined by

$$P_{2,j} = \frac{Q_j}{Q'_{\mathrm{t}}}, \qquad \sum P_{2,j} = 1, \tag{5}$$

$$P_{2,1} \le P_{2,2} \le P_{2,j} \le P_{2,n},$$

$$P_{2,1} + P_{2,2} + P_{2,j-1} \le R_2 \le P_{2,1} + P_{2,2} + \dots + P_{2,j},$$

where Q_j is the cross section for the process j and Q'_{t} is the effective total cross section after introduction of the "null process". The total collision cross section is defined as

$$Q_{\mathrm{t}} = Q_{\mathrm{el}} + Q_{\mathrm{a}} + Q_{\mathrm{v}} + Q_{\mathrm{ex}} + Q_{\mathrm{io}}. \tag{6}$$

Q_{el} is the elastic cross section, Q_{a} is the attachment cross section, Q_{ex} the electronic excitation cross section, Q_{v} the vibrational cross section and Q_{io} is the total ionization cross section. The sum of the fractional probabilities is equal to unity, and the interval [0,1] is divided into segments with lengths corresponding to these fractional probabilities. A random number R_2 between 0 and 1 is generated, and the interval into which this random number falls, determines the type of collision that occurs. The new energy and direction after the collision depends upon the type of collision: for excitation, the excitation threshold energy ε is given by $\varepsilon = \varepsilon_0 - \varepsilon_{\mathrm{exc}}$, where $\varepsilon_{\mathrm{exc}}$ is the excitation threshold and ε_0 is the electron energy before collision. For ionization, the total energy before collision is divided between the primary (original) electron and the secondary electron created in the ionization collision. For elastic collisions, the new kinetic energy of the electron is calculated by

$$\varepsilon = \varepsilon_0 \big[1 - 2 \big(m/M \big) \big(1 - \cos \chi \big) \big] \tag{7}$$

which is deduced from the hard sphere model. χ is the scattering angle of the electron after collision, where m and M are the masses of electron and molecule of SF_6, respectively. After a collision, the angles are determined by isotropic distribution. Hence, after the event of a collision, if the probabilities of inelastic collisions fail, the collision is deemed to be elastic. If the electron is attached, it is lost from the swarm and its subsequent fate is ignored. All electrons in the swarm moving forward and backwards, including the electrons formed during the ionization process, are traced until the termination time or loss due to attachment. During the successive collisions, for every electron, certain information (velocity, position, etc.) is stored in order to calculate, from appropriate sampling methods, the transport coefficients.

3. *Results and discussion*

In the Monte Carlo technique, the electron trajectories are calculated and collisions of electrons with molecules in the gas are simulated. The swarm parameters are obtained after following seed electrons from initial conditions for a long distance or long period of time. One of the difficulties in using the Monte Carlo technique in a highly electron attaching gas such as SF_6 is that the initial electrons released at the cathode get lost due to the high electron attachment cross section of SF_6. A summary of all cross sections [16 – 19] of SF_6 employed is shown in Fig. 1.

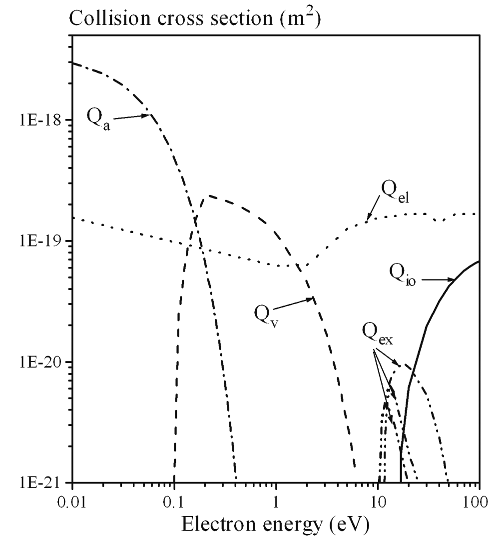
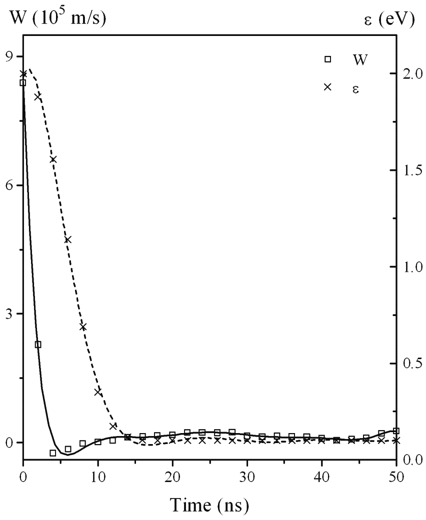


Fig. 1. Summary of collisional electron cross section of SF_6, Q_a – attachment, Q_v – vibrations, Q_{ex} – excitations, Q_{io} – ionization, Q_{el} – elastic.

The computing time for the Monte Carlo technique depends upon the number of test electrons released from the cathode and the number of collisions occurring while each electron travels the distance from the cathode to the anode. In order to achieve reliable values of the swarm parameters, simulations with 10000 test electrons are required, for zero field and low density – reduced electric field strength, E/N. The initial electrons are injected as a point source at $t = 0$ and $r = 0$ with a cosine distribution with a mean energy of 2 eV.

At low reduced electric field E/N values, in the case of electron molecule collisions, energies of electrons and target molecules are practically of the same order of magnitude. This is what happens at low electron energy not only during elastic collisions but also during inelastic collisions. At low E/N, there is another problem which can appear in the case of highly electronegative gases such as SF_6. The strong electron attachment occurring at low energies, can absorb enough initial seed electrons to stop Monte Carlo simulation. The purpose of this part is to present a Monte Carlo method available whatever E/N, but which is more adapted for low E/N values. Figures 2 and 3 show electron mean energy, drift velocity, and longitudinal and transverse diffusion coefficients under zero field conditions. Probably one of the most convincing validity tests of the treatment of low energy electron – molecule collisions with the Monte Carlo method is to determine distribution functions and transport coefficients under zero field conditions. Indeed, for an electron swarm or beam released (with known initial energetic and angular distributions) through a gas under zero electric field conditions, it is well established that this electron



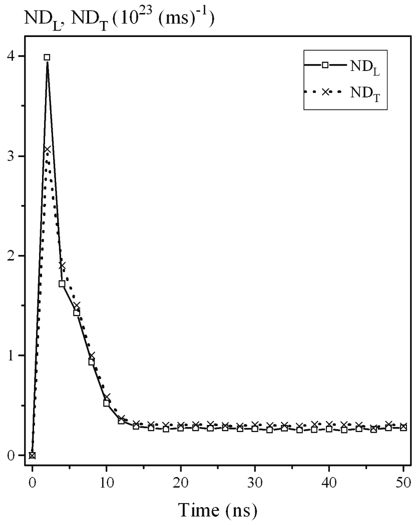


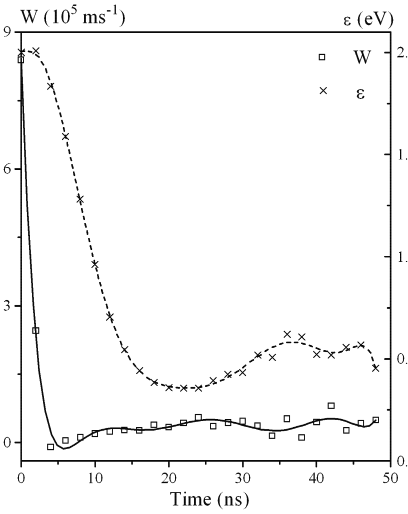
Fig. 2 (left). Zero field electron mean energy (ε) and drift velocity (W) as a function of time.

Fig. 3. Zero field longitudinal (ND_L) and transverse (ND_T) diffusion coefficients as a function of time.

swarm relaxes after a shorter or longer period of time (depending on initial conditions and background gas) towards an equilibrium distribution, whatever the initial distribution or the nature of the background gas. Such an equilibrium is obviously characterized by the classical behavior. The electron distribution function becomes Maxwellian at the background gas temperature, electron drift ceases and diffusion becomes completely isotropic (i.e., longitudinal and transverse diffusion coefficients are identical).

Figure 2 shows the drift velocity and an electron of mean energy which relaxes towards gas energy. Electrons emitted in the forward direction, after relatively few collisions, lose their initial anisotropic angular distribution so that the initial directed velocity becomes rapidly negligible. We often meet with a lack of swarm parameters of electrons, especially the longitudinal diffusion coefficient, when we attempt to simulate the plasma processing in reactive gases. Figure 3 shows the longitudinal and transverse diffusion coefficients. In this short time scale, the longitudinal diffusion coefficient, after an overshoot effect due to the anisotropy of the initial distribution, tends towards transverse diffusion. After a short time, the longitudinal and transverse diffusion coefficients decrease rapidly, this effects is due to the frequent vibrational collision and high attachment rates.

For $E/N = 10$ Td, the transport parameters are slightly higher than those for a zero field (see Figs. 4 and 5). The fluctuation of the transport coefficients in the first time period is attributed to the non-equilibrium in the electron energy distribution. The fluctuation in the latter is attributed to the statistical scatter since the number of electrons decrease rapidly due to the attachments. To reduce the scatter,



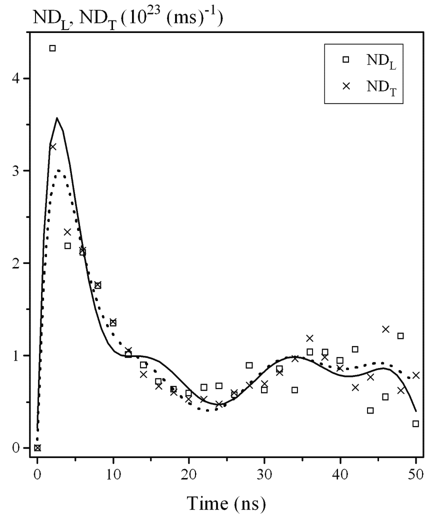


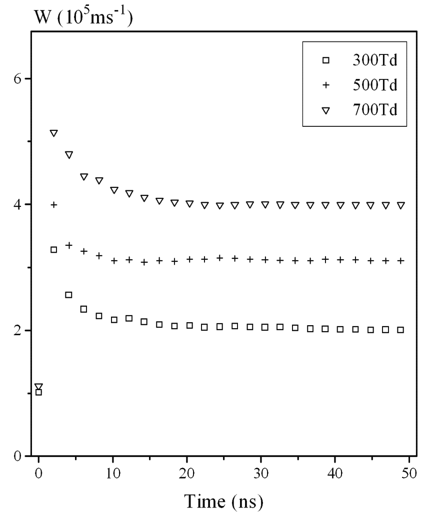
Fig. 4 (left). Electron mean energy (ε) and drift velocity (W) as a function of time, $E/N = 10$ Td.

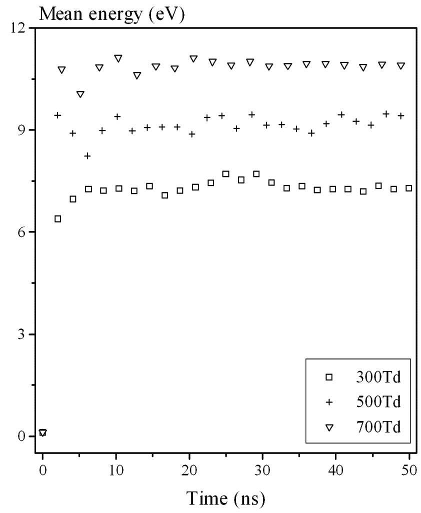
Fig. 5. Longitudinal (ND_L) and transverse (ND_T) diffusion coefficients as a function of time, $E/N = 10$ Td.

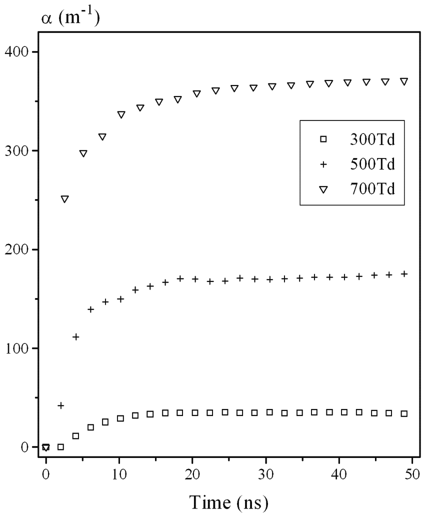
it was thought that a considerable number of electrons needs to be injected into the drift space in order to reduce the fluctuation. The Monte Carlo method is the preferred method at high values of E/N because it directly simulates the experimental method and also provides a check on whether the electrons have attained equilibrium. The computing time for the Monte Carlo technique depends on the number of test electrons released from the cathode and the number of collisions occurring while each electron travels a distance from the cathode to the anode.

Figures 6 – 9 show the variation of the drift velocity, electron mean energy, and ionization and attachment coefficients with time for different reduced electric field strength (E/N) values. The initial electrons are injected with a mean energy of 0.1 eV. This energy is low enough to not influence the behavior of the swarm at later times. A time is required for the drift velocity, electron mean energy and ionization coefficient to reach their steady states values. The electron attachment coefficient decreases with the time, due to the strong electron attachment cross section of SF_6 for low energy electrons. t_0 is the time required for the average electron energy to reach its steady state value.

In Figs. 10 and 11 showing the variation of α and the average electron energy ε with time. There is a time lag between the onset of steady state for the average energy and the onset of steady state for the ionization coefficient. The transient in α occurs because initially, $t \sim t_0$, the number of accumulated ionizing collisions is small, hence the ionization coefficient has not reached steady state. Figs. 10 and 11 also shows that the mean electron energy fluctuates, with diminishing amplitude of fluctuation, because the number of electrons in the avalanche is small in order







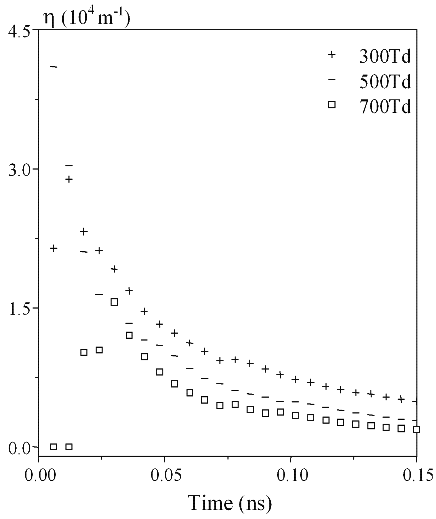


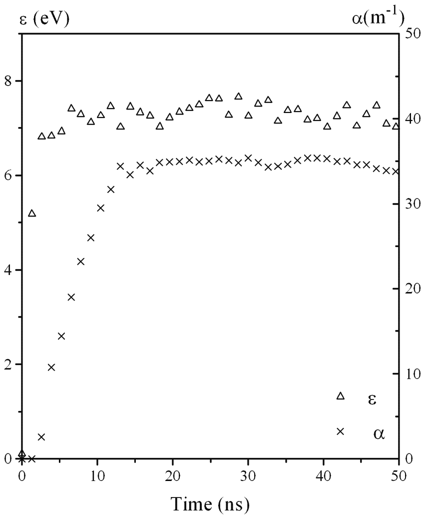
Fig. 6 (upper left). Electron drift velocity (W) as a function of time, for different E/N values.

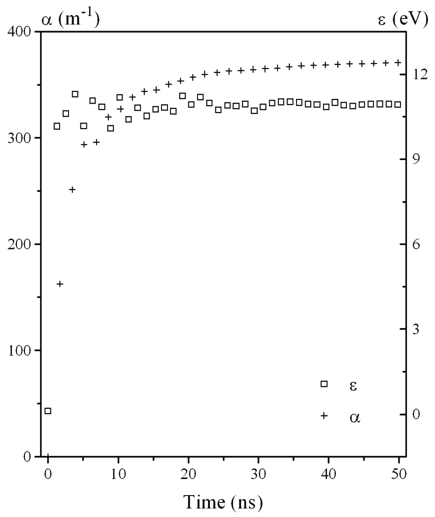
Fig. 7 (upper right). Electron mean energy as a function of time, for different E/N values.

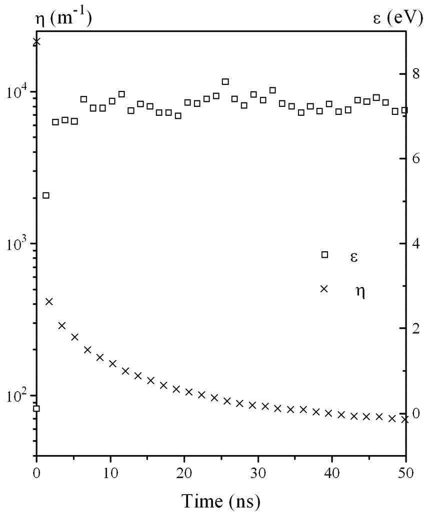
Fig. 8 (lower left). Electron ionization coefficient (α) as a function of time, for different E/N values.

Fig. 9 (lower right). Electron attachment coefficient (η) as a function of time, for different E/N values.

to reduce computational costs. The drift velocity, has the same qualitative time behavior as α, but reaches steady state in a shorter time.







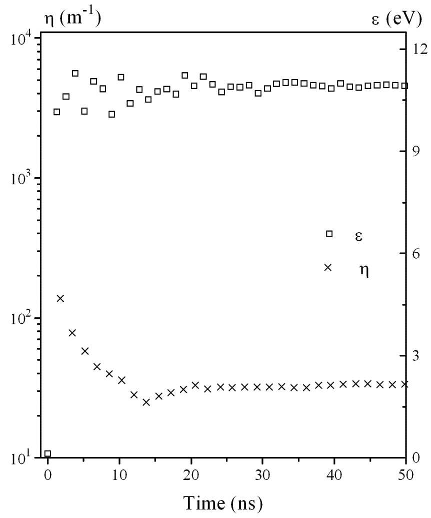


Fig. 10 (upper left). Mean energy ε and ionization coefficient α as a function of time, $E/N = 300$ Td.

Fig. 11 (upper right). Mean energy ε and ionization coefficient α as a function of time, $E/N = 700$ Td.

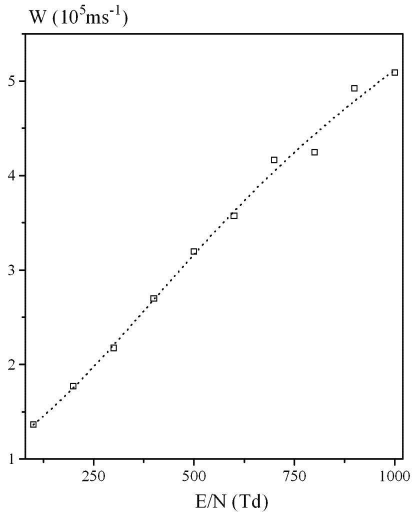
Fig. 12 (lower left). Mean energy ε and attachment coefficient η as a function of time, $E/N = 300$ Td.

Fig. 13 (lower right). Mean energy ε and attachment coefficient η as a function of time, $E/N = 700$ Td.

Figures. 12 and 13 show the variation of electron mean energy, and the attachment coefficient with time for different E/N values. There is a time lag between the onset of steady state for the average electron mean energy and the onset of steady state for the attachment coefficient. The attachment coefficient reaches the steady state in a shorter time with the increase of the reduced field. One notices, when using the Monte Carlo technique for a highly electron attaching gas such as SF_6, that the initial electrons released at the cathode get lost due to the high electron attachment cross section of SF_6.

There are fluctuations in the transport parameters depending on the distance from the cathode. In the low reduced electric field, after electronic excitation collision, electrons experience a few vibrational excitation collisions and disappear due to the electron attachment. The ionization coefficient increases and the attachment coefficient decreases with the increase of the reduced field.

As expected, statistical fluctuations are more pronounced in the case of the drift velocity, which is a statistical mean only of the component of the velocity along the z axis, as oposed to the mean energy value, which is an average of the sum squares of the three components of velocity, along x, y and z axes. Thus, for the same number of collisions, the electron mean energy is necessarily more accurate than the drift velocity.



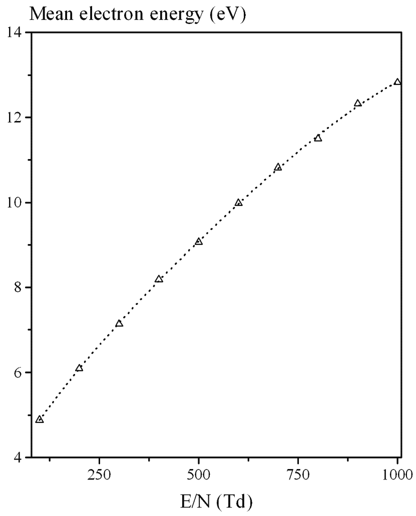


Fig. 14 (left). Electron drift velocity (W) as a function of E/N, fitted curve is the dashed line.

Fig. 15. Mean electron energy as a function of E/N, fitted curve is the dashed line.

Figures 14 and 15 show the variation of the electron drift velocity and the electron mean electron energy, as a functions of E/N. There is good agreement among the data for the electron drift velocity [20,21] from various experiments and the values calculated by the Monte Carlo method. A fairly good agreement is found between our values for the electron mean energy and those of various experiments [22 – 24].

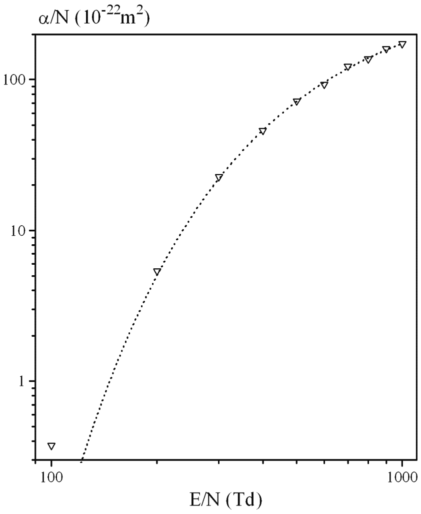
The drift velocities have been calculated with the Monte Carlo method and compared with the values calculated on the basis of the Boltzmann equation in the E/N range 30 − 550 Td [5]. The present results are about 20 % higher than those referred above at high reduced electric field. The electronic component constitutes only a small fraction of the total current collected at the anode, the attachment processes being dominant; most of the electrons emitted at the cathode are readily attached. Indeed, the dominance of an equilibrium region in the gap is not evident and may not warrant the hypotheses inherent in the approximation of the hydrodynamic regime in the Boltzmann analysis. The numerical solutions derived from the Monte Carlo method for the electron drift velocity in SF_6, have been compared with the result obtained in Refs. [25, 26]. The electron drift velocities reported in Ref. [25] were determined for SF_6 gas using the pulsed Townsend method for the reduced electric field values in the range 270 Td $\leq E/N \leq$ 360 Td, and over the range 32.24 Td $\leq E/N \leq$ 564.2 Td in Ref. [26]. Our data are generally higher than those measured in Refs. [25, 26] over the precedent studied range by approximately 15 % in the first range, and 25 % at high reduced electric field in the second range.

The experimental electron drift velocities determined using the voltage transient method [27] within the range 360 Td $< E/N <$ 720 Td agree quite well with the present results at the middle of the referred range, and fairly well in the low and high reduced electric field of this interval.

The present data agree well with the values of the electron drift velocity measured with the pulsed Townsend technique in Ref. [28] over the range of the reduced electric field strength E/N, from 50 to 360 Td. The data of the electron drift velocity measured by the pulsed Townsend technique [29] over the combined E/N range from 50 to 700 Td, are generally slightly lower than our calculated values. For comparison, we show the recently measured values of the drift velocity [6] with the pulsed Townsend technique over the reduced electric field strength E/N, between 100 and 700 Td. It is seen that, in the studied range, our values are slightly higher than in Ref. [6]. The plot of electron drift velocity as a function of E/N, in the range 200 to 1000 Td, shows a good agreement of the data for the electron drift velocity from experiments [17] with our values calculated by the Monte Carlo method.

The good agreement between experimental data and the simulation results shown in Figs. 14 and 15 indicates that the collision calculations accurately predict the growth of electron pulses into electron avalanches. The relevance of macroscopic coefficients, such as the first ionization coefficient, the attachment coefficient and the dielectric strength have long been recognized, but the description and prediction of dielectric behavior have traditionally been conceived in terms of empirical and semi-empirical formulae and/or simple criteria based, for example, on the molecular mass or the boiling point. Although these have proved useful for practical purposes, a more accurate and more general description is needed in the context of new gaseous insulators. Such a description is based on micro-physical material properties, collisional cross sections. Its primary advantage is to provide a basis for qualitative prediction of the insulating properties of gases and gaseous mixtures.

In view of the practical importance to the engineers, the swarm parameters α/N (reduced ionization coefficient) and η/N (electron attachment coefficient divided by the gas number density) generated by the simulation technique in SF_6 are shown in Figs. 16 and 17. The attachment coefficient is a measure of the probability that an electron will attach to a gas molecule in traveling a unit distance in the electric field direction.



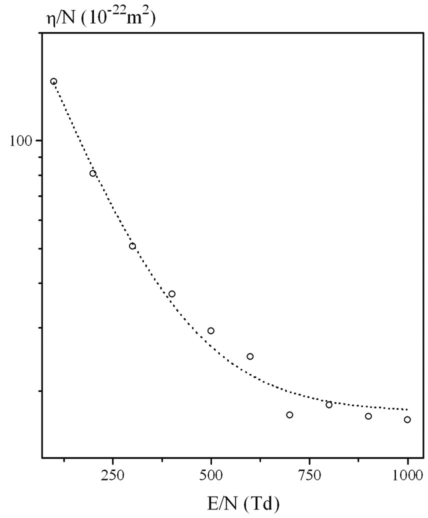


Fig. 16 (left). Reduced ionization coefficient α/N as a function of E/N, curve from Eq. (8) is the dashed line.

Fig. 17. Reduced electron attachment coefficient η/N as a function of E/N, fitted curve is the dashed line.

The reduced electron attachment coefficient decreases when increasing the normalized electric field E/N. Figure 17 shows that at low reduced electric field, the SF_6 gas has very large attachment coefficient. Therefore, most electrons, after a few free flights, can be attached. The reduced attachment coefficient becomes small for high values of the reduced electric field due to the large ionization cross section of SF_6.

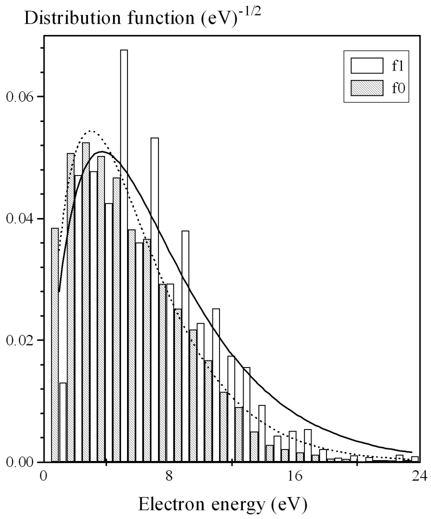
The inelastic collisions reduce electron energy and improve dielectric strength. The reduced ionization coefficient (α/N) is related to E/N according to the semi-empirical equation

$$\alpha/N = A \exp\{-B/(E/N)\} \tag{8}$$

where A and B are constants characteristic of the gas.

There has been a number of measurements of the ionization coefficient. In general, the calculated data of the attachment coefficients with the Monte Carlo technique agree well with the experimental results [20,22,24,30 − 32].

Here, we confirm that the Monte Carlo method is valid for deduction of the swarm parameters at low, intermediate and high reduced electric field E/N values despite the fact that SF_6 is a strongly electronegative gas. For a set of n particles and a given simulation time, the particle energy distribution function (or electron energy distribution function) can be determined. To obtain good statistics, a large number of particles are required. The electron energy distribution at $E/N = 200$ and 400 Td are shown in Figs. 18 and 19. The anisotropic parts of the distribution are quite small in the case of $E/N = 200$ Td as shown in Fig. 18. By contrast, the distribution at $E/N = 400$ Td shows much greater anisotropic parts than the isotropic part alone. This implies that the low E/N fluctuation results from the acceleration of each electron during the interval between collisions in the low energy region where the direction of motion of electrons easily changes by the electric field. The dot and full lines show the Maxwellian distribution at the same energy. The agreement between the present calculated distribution function and those results obtained by Yoshizawa et al. [33] is good.



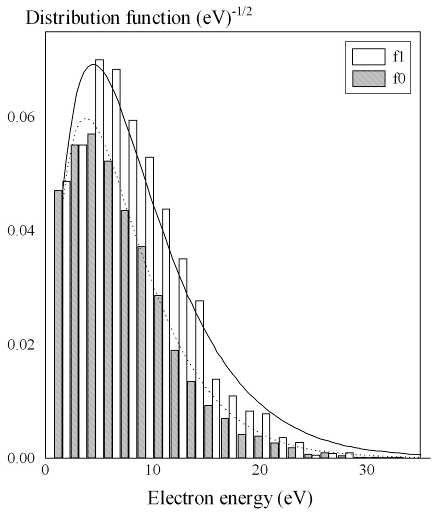


Fig. 18 (left). Electron energy distribution at $E/N = 200$ Td*: $f0$ – isotopic part, $f1$ – with anisotropic parts.*

Fig. 19. Electron energy distribution at $E/N = 400$ Td*: $f0$ – isotopic part, $f1$ – with anisotropic parts.*

4. *Conclusion*

Studies of SF_6 have been motivated by the importance of this gas for plasma etching of metals and silicon, for negative ion sources and in development of gaseous dielectrics. Monte Carlo simulations have become increasingly important as a numerical tool, particularly in the area of low-temperature plasma physics. In this study, we have examined the behavior of electrons in uniform electric fields using the Monte Carlo simulation. Electron swarm parameters have been calculated as a function of reduced electric fields E/N. Binary electron – neutral gas molecule collisions are the essential mechanism in the electron avalanche growth. The simulation results give values for electron drift velocity, electron mean energy, ionization and attachment coefficients, longitudinal and transverse diffusion coefficient, and electron energy distribution as functions of time and reduced electric fields. The good agreement between calculated and measured swarm parameters demonstrates the validity of the binary collision simulation techniques. A study with large number of electrons is needed to obtain stable values of the coefficients with high resolution at low values of E/N. Energy distributions obtained by the simulation indicate Maxwellian tail behaviors at corresponding mean energies.

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OPONAŠANJE ELEKTRONSKIH ROJNIH PARAMETARA U SF_6

Oponašamo gibanje elektrona u sumpornom heksafluoridu u jednolikom električnom polju primjenom Monte Carlo metode. Uspoređujemo određene rojne parametre s eksperimentalnim rezultatima za posmičnu brzinu, srednju energiju elektrona, te omjere ionizacijskog i veznog koeficijenta s brojevnom gustoćom molekula. Usvojeni udarni presjeci za sudare elektron – molekula za račune oponašanja u dobrom su skladu s eksperimentalnim vrijednostima u promatranom području E/N (E je jakost električnog polja a N brojevna gustoća molekula plina).