International Journal of Aquatic Science
ISSN: 2008-8019
Vol 12, Issue 02, 2021

Electron Transport Coefficients ForCHF_3

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Abstract: The two-term solution of Boltzmann equation has been used to evaluated the electron energy distribution function (EEDF) and electron transport parameters in pure electronegative gas CHF_3 for a wide range of E/N varying from 0.1 to 200 Td (1 Td=10^{-17} V.cm^2). This parameters, are electron drift velocity, mean electron energy, characteristic energy, electron temperature, ionization coefficient, attachment coefficient, effective ionization coefficient and reduced critical electric field strength $(E/N)_{cr}$. The present calculated results are in good agreements as compared with the previous experimental and theoretical results. A set of elastic and inelastic cross-sectionare collected for CHF_3 gas to evaluated transport parameters over the entire E/N range.

Keywords: CHF_3 molecule, EEDF, limiting field strength, electron swarm parameters, Boltzmann equation, cross section.

1. INTRODUCTION

Trifluoromethan (CHF_3) known as fluoroform, a kind of clean hydro-fluorocarbon extinguishing gas, is a replacement of CF_4 and SF_6 due to its zero ozone depletion potential, relative low toxicity, insulation and low boiling point[1]. CHF_3 is used in various application such as, polystyrene industries, refrigerant industrial, manufacture of Teflon, in the semiconductor industry for etching and deposition of silicon compounds [2,3], it is a good candidate for silicon dioxide (SiO_2) and silicon nitride thin film by using plasma enhanced chemical vapor deposition (PECVD),all of which result in emissions to the atmosphere. It is reported by McCulloch and Lindley [2], andOram, et al., [3]since 1978 the rate concentration of CHF_3 in the atmosphere increases by 5% per year. However, CHF_3 is a good greenhouse gas with a global warming potential (GWP) is nearly two times higher than that of CF_4, its usage is limited by Kyoto Protocol[6]. CHF_3 and CF_4 are both important in low pressure plasma processing gas with the global warming potential of CHF_3 and CF_4 are 11700 and 6300 times greater than that of CO_2 for a 100 year period, only 50.63% and 26.36% of SF_6 respectively [7,8], and the atmospheric life-time of the gas is 264 years less than that of CF_4[9]. In order to decrease the greenhouse of the buffer gas, CHF_3 involved mixtures with strongly electron attaching gases for gaseous dielectrics purposes have been used in electrical power engineering.

Generally, Trifluoromethan molecule is polar buffer gas its electric dipole moment is $5.504x10^{-30}$C.m (1.65 debye)[10], at room temperature and atmospheric pressure, is a colorless, odorless, non-flammable , non-explosive characteristics, heavier than air with density 0.673 g/cm^{3}at 298 K and vapor pressure 4.38 MPa at 293 K, may asphyxiate by displacing air. CHF_3 has been used not only in the form of pure gas but also in mixtures with other gases (such as SF_6, CF_4 and c-C_4F_8 mixture gas) for much industrial application.Research works of de Urquijo, et al., [11] and de Urquijo, et al., [12]have

revealed that the breakdown voltage for pure CHF_3 nearly equal to 80Td which is lower that the uniform field breakdown voltage ~97.5 Td studied by Christophorou, et al.,[13], and lower than that of pure SF_6.

Since the original meaning of swarm is close to a "group", the behaviors concerning to the motion of electrons and ions in plasma or neutral species under the effect of an external uniform dc electric field can be treated in terms of the swarm.However, Takeda, [14] investigated that most of the theoretical and experimental swarm analyses are independent of the electron density. Even though the electron energy distribution function EEDF play important role in plasma processing and gas discharge phenomena and then electron swarm parameters are calculated using the EEDF. In thermal equilibrium the EEDF has a Maxwellian shape, if the external electric field increase, the EEDF slightly deviates from the strict Maxwellian one.

The electron swarm parameters in pure CHF_3 gas which are the electron drift velocity, diffusion coefficient, and effective ionization coefficients, are experimentally reported in the literature [11, 13, 15-17] using a pulsed Townsend technique, and a double shutter drift tube with variable drift distance [18].Theoretically the electron swarm parameters of $CHF_3$are calculatedfrom the estimated cross sections by the Boltzmann equation analysis [8,19], and Monte Carlo method [20,57-61].These swarm parameters, are also calculated in binary CHF_3-CF_4 gas mixtures [21] and in CHF_3-Ar Gas mixtures [20], and its mixtures with Ar, SF_6 and N_2 are also investigated experimentally[16,22-24]. Furthermore, the ternary mixtures CHF_3-Ar-O_2 were also analyzed by Kim, et al., [25].

In this study, we calculate the EEDF, and the electron swarm parameters which are, the electron drift velocity, mean electron energy, characteristic energy, electron mobility, diffusion coefficient, ionization, attachment and effective ionization coefficients, by solving Boltzmann equation with two-term expansion for steady Townsend discharge, in the E/N rang of 0.1 -1000 Td, (1 Td=10^{-17} V.cm^2) using NOMAD code [26, 48-56]. The reduced critical electric field strength $(E/N)_{cr.}$ is calculated at which reduced ionization coefficient exactly balances with reduced attachment coefficient,where E is electric field and N is the gas density.

2. BOLTZMANN EQUATION

In the case of weakly ionized gas, the electron energy distribution function EEDF can be calculated by using the Boltzmann equation [27-29]

$$\frac{\partial f}{\partial t}+\bar{v}.\nabla_r f-\frac{e\bar{E}}{m}.\nabla_v.f=\left(\frac{\partial f}{\partial t}\right)_C \qquad (1)$$

where $f=f\left(\bar{r},\bar{v},t\right)$is the electron of positions $\bar{r}$ and electron velocity $\bar{v}$, a=e$\bar{E}$/m is the acceleration due to electron field $\bar{E}$, e is the electron charge and m is the electron mass, ($\partial f/\partial t)_c$ is the collision operators which is the rate change of the number of electrons per unit volume of phase space [30].

For a specially uniform gas, so that the velocity distribution function is function of velocity only, $f\left(\bar{r},\bar{v},t\right)=f\left(,\bar{v},t\right)$, where $\nabla_r f=0$, the Boltzmann equation for electrons is,

$$\frac{\partial f}{\partial t}-\frac{e\bar{E}}{m}.\nabla_v.f=\left(\frac{\partial f}{\partial t}\right)_C \qquad (2)$$

The electron velocity distribution function is spherically symmetric, if the electron collision frequency for elastic collision is larger than the electron collision frequency for inelastic

collision, so the two-terms expansion of the $f(\bar{r},\bar{v},t)$ in the zero-order Legendre Polynomial $P_J(\cos\theta)$ will be sufficient[31-33],thus $f(\bar{v})$takes the form,

$$f(,\bar{v},)= f_o(v)+\frac{\bar{v}}{v} f_1(v) \qquad (3)$$

Where $f_o(v)$ and $f_1(v)$ represented the isotropic and anisotropic part, respectively, are function of velocity only, and $f_1(v) << f_o(v)$.

Substituting Eq. (3) into Eq. (2), we can get the basic equation of the Boltzmann equation in the Lorentz approximation as given by Frost and Phelps, [34], without superelastic (second kind) collision[35],

$$\frac{d}{d\varepsilon}\left(\frac{e^2E^2\varepsilon}{3NQ_m(\varepsilon)}\frac{df_0(\varepsilon)}{d\varepsilon}\right)+\frac{2m}{M}\frac{d}{d\varepsilon}\left[\varepsilon^2 NQ_m(\varepsilon)\left\{f_o(\varepsilon)+K_BT_g\frac{df_o(\varepsilon)}{d\varepsilon}\right\}\right] \qquad (4)$$

$$\sum_J(\varepsilon+\varepsilon_J)f_0(\varepsilon+\varepsilon_J)NQ_J(\varepsilon+\varepsilon_J)-\varepsilon f_0(\varepsilon)N_J\sum_J Q_J(\varepsilon)=0$$

Where $f_o(\varepsilon)$ is the electron energy distribution function EEDF, m/M is the ratio of electronic to atomic mass, T_g is the gas temperature, K_B is the Boltzmann constant, $Q_m(\varepsilon)$ is the momentum transfer cross-section, $Q_J(\varepsilon)$ is the cross-section in which the electron loss energyε_J during excitation to the J^{th} level. Thus, $\varepsilon=mv^2/2e$, ε is the electron energy in electron volt (eV).

In the case of thermal-equilibrium plasma at relative low reduced electric field strength E/N, the EEDF is normally Maxwellian[36],

$$f_o(\varepsilon)=\left(2/\sqrt{\pi}\right)\exp(-\varepsilon/K_BT)/(K_BT)^{3/2} \qquad (5)$$

Would give the ratio of transverse diffusion coefficient to electron mobility D_T/μ_e equal to 2/3 the mean electron energy $\langle\varepsilon\rangle$. In fact in the E/N range concerned, $D_T/\mu_e > \langle\varepsilon\rangle$, theelectron energy distribution function is strongly non-Maxwellian.

3. TRANSPORT PARAMETERS

The electron swarm parameters directly depend on EEDF, and all of the collision cross-sections called the total effective collision cross-section for momentum transfer $Q_m^T(\varepsilon)$ which is given by,

$$Q_m^T(\varepsilon)=Q_m(\varepsilon)+Q_i(\varepsilon)+Q_a(\varepsilon)+\sum_J Q_{ex}(\varepsilon) \qquad (6)$$

Where $Q_m(\varepsilon)$, $Q_i(\varepsilon)$, $Q_a(\varepsilon)$ and $Q_{ex}(\varepsilon)$, denote the electron cross-sections of momentum transfer, ionization, attachment, and excitation (vibration, electronic) respectively. For a gas mixture,

$$Q_m^T(\varepsilon)=\sum_n K_nQ_{m,n}^T \qquad (7)$$

Where $K_n$is the ratio of gas in binary mixture of gases[37].

The electron energy distribution function $f(\varepsilon,E/N)$, is normalized by,

$$\int_0^\infty \sqrt{\varepsilon}\,f(\varepsilon,E/N)d\varepsilon=1 \qquad (8)$$

Once the electron energy distribution function is found, then the electron swarm parameters can be calculated.

The electron drift velocityand mean electron energy were calculated as follows [38-41],

$$v_d = -\frac{\gamma E}{3N}\int_0^\infty \frac{\varepsilon}{Q_m^T(\varepsilon)}\frac{\partial f_0(\varepsilon)}{\partial \varepsilon}d\varepsilon \qquad (9)$$

$$\langle \varepsilon \rangle = \int_0^\infty \varepsilon^{3/2} f_0(\varepsilon) d\varepsilon \qquad (10)$$

The reduced electron mobility, reduced transverse diffusion coefficient and characteristic energy are calculated from[36,42,43],

$$\mu_e N = -\frac{\gamma}{3}\int_0^\infty \frac{\varepsilon}{Q_m^T(\varepsilon)}\frac{\partial f_0(\varepsilon)}{\partial \varepsilon}d\varepsilon \qquad (11)$$

$$D_T N = \frac{\gamma}{3}\int_0^\infty \frac{\varepsilon}{Q_m^T(\varepsilon)} f_0(\varepsilon) d\varepsilon \qquad (12)$$

$$\varepsilon_k = \frac{eD_T}{\mu_e} \qquad (13)$$

Where $\gamma = (2e/m)^{1/2}$ is a constant, and N is the concentration of gas calculated using the equation,

$$N = \frac{\rho N_A}{M} \qquad (14)$$

Where ρ is the gas density, M is molecular weight, and N_A is the Avogadro number.

The ionization, attachment and effective ionization coefficients are obtained as follows [44-46],

$$\frac{\alpha}{N} = \frac{\gamma}{v_d}\int_i^\infty Q_i(\varepsilon) f_0(\varepsilon) \varepsilon d\varepsilon \qquad (15)$$

$$\frac{\eta}{N} = \frac{\gamma}{v_d}\int_a^\infty Q_a(\varepsilon) f_0(\varepsilon) \varepsilon d\varepsilon \qquad (16)$$

$$\frac{\alpha - \eta}{N} = 0 \qquad (17)$$

Where, $Q_i(\varepsilon)$, $Q_a(\varepsilon)$ are ionization and attachment cross section, here, i and a is the thresholdenergy. The reduced critical electric field strength $(E/N)_{cr.}$is calculated from Eq. (17), when the formation and loss electrons reach a balance[47].

4. CHF_3 CROSS-SECTION

The calculation of the electron swarm parameters and critical field strength requires different kinds of elastic and inelastic collision processes. The cross-section set of CHF_3has been previously discussed and estimated by [8,17,19,48]. In the present study, the momentum transfer cross-section was taken from Voloshin, et al., [48]. Three vibrational electron impact cross-sections (v_{14}, v_{25}, and v_{36})with threshold energy 0.37, 0.18, and 0.13 respectively, were considered taken from Kushner and Zhang [17]. The suggested ionization and attachment cross section set with threshold energy 14.02 eV and 10.0 eV respectively, by Morgan, [19] are used without any modification.

5. RESULTS AND DISCUSSION

A set of electron collision cross-sections explained in section 3 has been used in present calculation to obtain electron swarm parameters and comparison with the previous experimental and theoretical swarm data. The energy range of cross-sections has been taken up to 100eV, which is typical of the breakdown voltage.

Electron energy distribution function EEDF and electron swarm parameters were calculated for a typical das number density N=2.4615x10^{19} cm^{-3}, which is equivalent to 1 atm at 298 K. Figure 1 shows the effect of the reduced electric field strength on the EEDF in CHF_3. The energy variation of cross sections has a very majoreffect on the electron energy distribution function for typical conditions of electric discharges.At lower energies E/N<20 Td, and below the threshold of excitation of the vibrational state, whose its threshold energy is equal to 0.13eV, the normalized distribution appear close to the Maxwellian distribution function, in this region the EEDF decrease with increasing E/N. However, for electron energies greater than the 0.13 eV the EEDF increase with increasing E/N, at high E/N the inelastic collision processes are dominate the distribution which is being non-Maxwellian.

Figure 2 are shown the influence of superelastic (second kind) collisions on EEDF at reduced electric field strength 1Td and 50Td with and without superelastic collision respectively. At low reduced electric field strength E/N, the effect of superelastic collision on EEDF are important. The study of the influence of superelastic collisions was explained byPietanza, et al., [49].

Figure 3 shows the electron drift velocities as function of the E/N range from 0.1 to 1000 Td for CHF_3. At low reduced electric field E/N, the electrons are in thermal equilibrium, the value of drift velocity for pure CHF_3 are low, due to the large scattering cross-section caused by the large electric dipole moment (5.504x10^{-30}C.m) [10]. The present electron drift velocity is in good agreement with the experimental results [16,17,24,50,51] and theoretical results [21], are also plotted in same figure.

Figure 4 shows the mean electron energy as a function of E/N, in the low electric field strength E/N>10 Td the mean energy is in thermal equilibrium, it is essentially isotropic remain nearly constant, at high E/N the mean electron energyrapidly increases with increasing E/N, because its sensitive to inelastic collisions. The behavior of the mean electron energy is also reflected in the electron drift velocity and diffusion coefficient. The present calculation was found in good agreement with theoretical values [21].

Figure 5 shows calculated electron temperature T_e as a function of E/N, from the relation $\langle \varepsilon \rangle = (3/2)T_e$, where $\langle \varepsilon \rangle$ is mean electron energy and T_e is electron temperature in unit eV. The variation in electron temperature is more nonlinear at E/N >10 Td, this due to inelastic cross sections of CHF_3 molecule. The experimental values of Kushner and Zhang, [17] are lower over the reduced electric field E/N range greater than 40 Td, when compared with the present calculation. The characteristic energy in pure CHF_3 as a function of E/N is shown in figure 6, at the low electric field strength E/N>10 Td the characteristic energy is in thermal equilibrium, at high E/N the characteristic energy increases with increasing E/N, because the inelastic collisions are dominate.

The reduced transverse diffusion coefficient calculated in this work for $CHF_3$is shown in figure 7. The experimental value ofXiao and Deng [51] was also included which show a good agreement with present work.

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Figure 8 shows the reduced ionization coefficient α/N, has been calculated for the rang 40 ≤ E/N ≤ 1000 Td. The ionization coefficient increase as E/N increases, good agreement has been obtained with the theoretical results [21] and the experimental results [11,24]. Ionization coefficient appear to be in good agreement in the region of critical reduced field strength $(E/N)_{crt}$. The attachment coefficient, is the probability that an electron will attach with the molecule in traveling a unit distance in electric field, is only a function of E/N. The calculated reduced, attachment coefficient, η/N for the pure CHF_3 molecule as a function of E/N is shown in figure 9. The present calculation was found to be in good agreement, with theoretical results of Duzkaya and Tezcan, [21].

The reduced effective ionization coefficient in CHF_3 is shown in figure 10. It can be seen from this figure that at the limiting field strength$(E/N)_{lim}$is about 78 Td, the effective ionization coefficient increases with increasing *E/N* where the ionization collisions become. There is a good agreement with the theoretical results of Duzkaya and Tezcan[21]and experimental results of Christophorou and Olthoff [8] over all the E/Nrange covered in the present study.

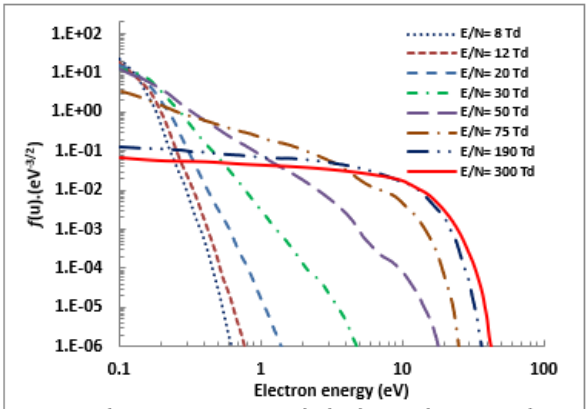

Figure 1. Electron energy distribution function as a function of electron energy at different values of E/N.

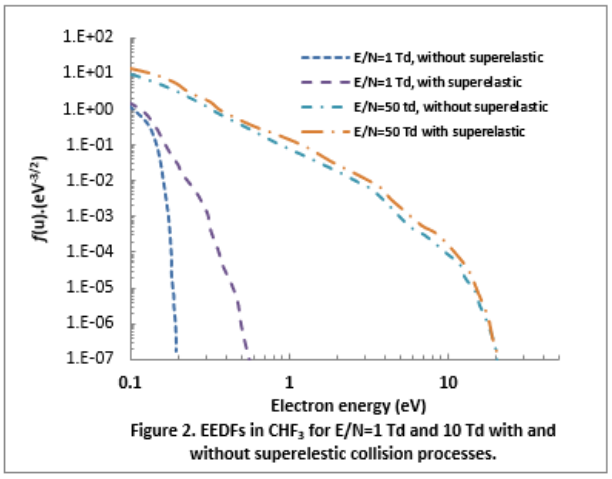

Figure 2. EEDFs in CHF_3 for E/N=1 Td and 10 Td with and without superelestic collision processes.

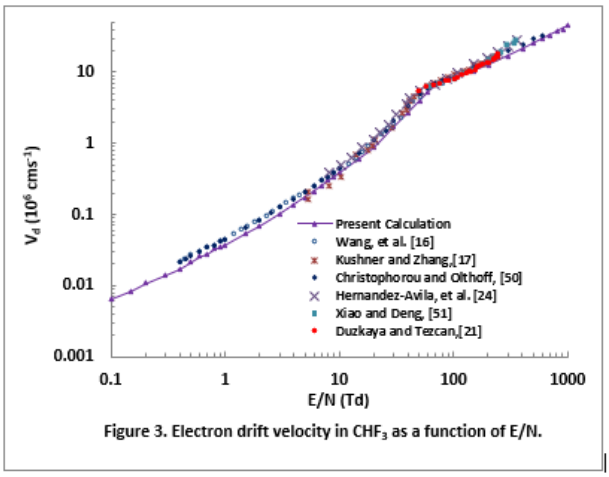

Figure 3. Electron drift velocity in CHF_3 as a function of E/N.

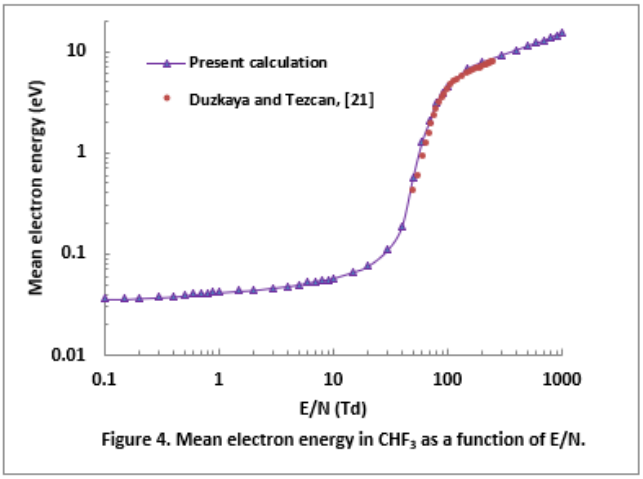

Figure 4. Mean electron energy in CHF_3 as a function of E/N.

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ISSN: 2008-8019
Vol 12, Issue 02, 2021

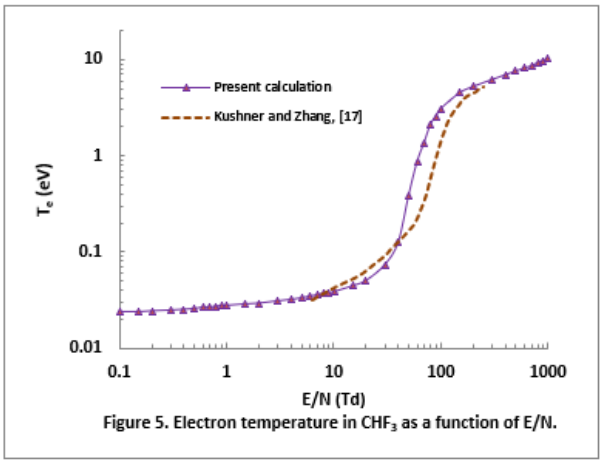

Figure 5. Electron temperature in CHF_3 as a function of E/N.

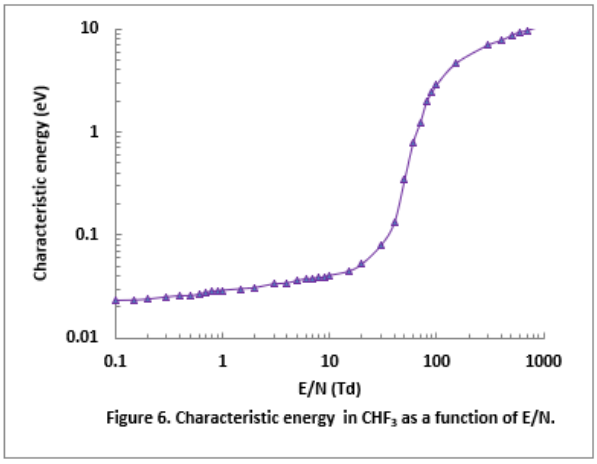

Figure 6. Characteristic energy in CHF_3 as a function of E/N.

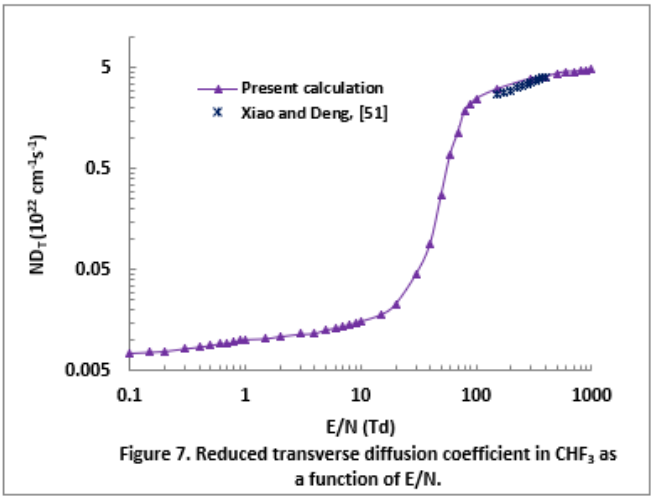

Figure 7. Reduced transverse diffusion coefficient in CHF_3 as a function of E/N.

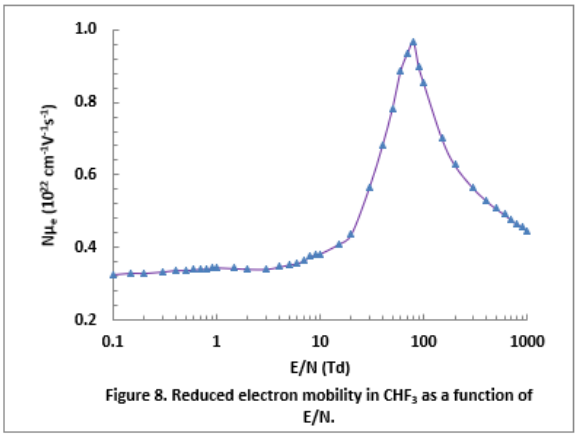

Figure 8. Reduced electron mobility in CHF_3 as a function of E/N.

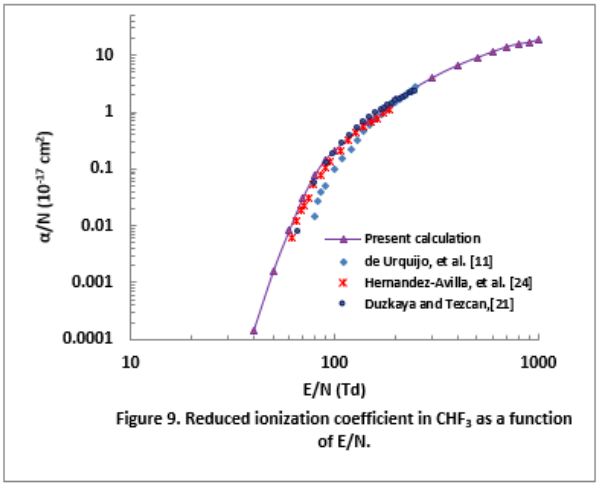

Figure 9. Reduced ionization coefficient in CHF_3 as a function of E/N.

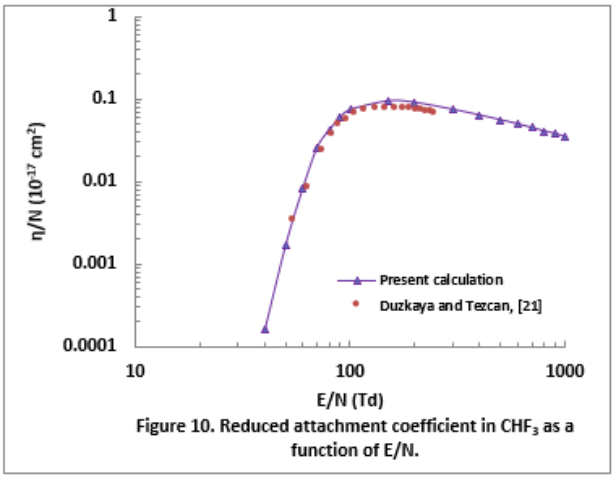

Figure 10. Reduced attachment coefficient in CHF_3 as a function of E/N.

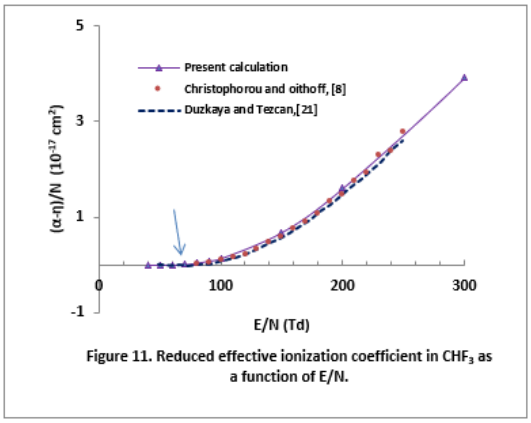

Figure 11. Reduced effective ionization coefficient in CHF_3 as a function of E/N.

6. CONCLUSION

For CHF_3 a set of collision cross-section have been used as input data with the two-term solution of Boltzmann equation method, will accurately predict the values of the electron swarm parameters, and limiting field strength are calculated over a wide range of E/N, varying from 0.1 Td to 1000 Td. In comparison present calculation is found to be in good agreement with previous theoretical and experimental values.

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