

Calculation of Electron Swarm Parameters in Tetrafluoromethane

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Abstract—The electron swarm parameters and electron energy distribution function (EEDF) are necessary, especially on understanding quantitatively plasma phenomena and ionized gases. The EEDF and electron swarm parameters including the reduce effective ionization coefficient $(\alpha-\eta)/N$ (α and η are the ionization and attachment coefficient, respectively), electron drift velocity, electron mean energy, characteristic energy, density normalized longitudinal diffusion coefficient, and density normalized electron mobility in tetrafluoromethane (CF_4) which was analyzed and calculated using the two-term approximation of the Boltzmann equation method at room temperature, over a range of the reduced electric field strength (E/N) between 0.1 and 1000 Td ($1\text{Td}=10^{-17}\text{V.cm}^2$), where E is the electric field and N is the gas density of the gas. The calculations required cross-sections of the electron beam, thus published momentum transfer, vibration, electronic excitation, ionization, and attachment cross-sections for CF_4 were used, the results of the Boltzmann equation in a good agreement with experimental and theoretical values over the entire range of E/N . In all cases, negative differential conductivity regions were found. It is found that the calculated EEDF closes to Maxwellian distribution and decreases sharply at low E/N . The low energy part of EEDF flats and the high-energy tail of EEDF increases with increase E/N . The EEDF found to be non-Maxwellian when the $E/N > 10\text{Td}$, having energy variations which reflect electron/molecule energy exchange processes. In addition, limiting field strength $(E/N)_{\text{limit}}$ has been calculated from the plots of $(\alpha-\eta)/N$, for which the ionization exactly balances the electron attachment, which is valid for the analysis of insulation characteristics and application to power equipment.

Index Terms—Boltzmann equation, CF_4 , Electron discharge, Electron swarm parameters, Kinetic and transport theory.

I. INTRODUCTION

Tetrafluoromethane, also known as carbon tetrafluoride or R-14 is the simplest fluorocarbon (CF_4), was first observed

as an impurity in krypton by Gassman, 1974, it has a low critical temperature and high critical pressure. The CF_4 is a man-made gas which has an important role in technological applications such as semiconductor, etching plasma for material processing, discharge opening switches, atmospheric physics and chemistry, gaseous dielectrics, and gaseous detector technology (Hunter, et al., 1985; Proshina, et al., 2015). Tetrafluoromethane is combined with oxygen as an etching medium used in the semiconductor industry for etching of dielectric materials, such as SiO_2 and also for deposition of fluorinated polymer films. In combination CF_4 with SF_6 produce a less expensive, toxic, and corrosive cryogenic liquid than SF_6 alone (Reinking, et al., 1986).

Tetrafluoromethane (CF_4) is a very powerful greenhouse, it is stable and does not deplete the ozone layer, which strongly absorbs infrared radiation at $\sim 8\ \mu\text{m}$. The lifetime of CF_4 is 50,000 years, with a global warming potential (GWP) 6500 times greater than that of CO_2 over a 100-year time scale, only about 26.36% of that of SF_6 (Muhle, et al., 2010; Tezcan, et al., 2016).

Tetrafluoromethane, at room temperature and atmospheric pressure, is a colorless, odorless, easy to compress gas, high stability, non-toxic, and non-flammable gas. CF_4 has been used not only in the form of pure but also in mixtures with other gases for much industrial application. In high-voltage technology, CF_4 is used as an admixture for circuit breakers (Duzkaya and Tezcan, 2017).

For example, binary mixtures of CF_4 and Ar are used in microelectronics circuit applications (Cox, et al., 1989), whereas CF_4/Ar mixtures have been also used in the application of capacitively coupled plasma (CCP) (Kitajima, et al., 2000) and possibly even inductively coupled plasma (Hioki, et al., 2000). The mixture of SF_6 and CF_4 has found applications in high-voltage circuit breakers in a cold climate which has greatly reduced the GWP compared with pure SF_6 (Xiao, et al., 2004). A mixture of 50% SF_6 –50% CF_4 has been proposed as an insulating gas for electrical equipment, which was successfully utilized for circuit breakers (Wu, et al., 2006).

The electron swarm parameters of CF_4 , which are the drift velocity, characteristic energy, mean electron energy, electron mobility, diffusion coefficient ionization, attachment, and effective ionization coefficients, are widely investigated in the

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literature (Bordage, et al., 1996; Bordage and Segur, 1999; Vasenkov, 1999; Christophorou and Olthoff, 2004; Xiao and Deng, 2013; Sang-Nam, 2015; Michele, 2018). These swarm parameters are also calculated in CF₄-Ar (Kurihara, et al., 2000; Tezcan, et al., 2013), CF₄-SF₆ (Xueli and Xiao, 2007), and CHF₃-CF₄ gas mixtures (Duzkaya and Tezcan, 2019). Furthermore, the ternary mixtures SF₆-CF₄-Ar were also analyzed by solving Boltzmann's equation (Tezcan, et al., 2016; Tezcan, et al., 2016).

In the present study, the behavior of electron swarm parameters in pure tetrafluoromethane (CF₄) is studied and analyzed using the two-term approximation of the Boltzmann equation in the range of density reduced electric field strength E/N varying from 0.1 to 1000 Td (1Td=10⁻¹⁷ Vcm²), where E is the electric field and N is neutral number density.

II. THEORY

A. The Boltzmann Equation

The role of electron energy distribution function (EEDF) is an important parameter to compute the electron swarm parameters, it is difficult to measure experimentally. The EEDF calculated from a set of electron gas collision cross-section using Monte Carlo simulation method or Boltzmann equation (two-term or multi-term solution), when the electron collision frequency for inelastic collisions is much smaller than the collision frequency for elastic collision, it is the condition the two-term solution to be valid (Smith and Thomson, 1978). The steady-state distribution function is then given by the solution of the Boltzmann equation which is based on the two-term theory (Pinheiro and Loureiro, 2002). A detailed solution of Boltzmann equation used in this study to calculate electron swarm parameters may be found in the literatures (Holstein, 1946; Frost and Phelps, 1962; Smith and Thomson, 1978; Hagelaar and Pitchford, 2005). The basic of Boltzmann transport equation used in this study is given as,

$$\begin{aligned} & \frac{E^2}{3} \frac{d}{d\varepsilon} \left(\frac{\varepsilon}{NQ_m^T(\varepsilon)} \frac{df_0(\varepsilon)}{d\varepsilon} \right) + \frac{2m}{M} \frac{d}{d\varepsilon} \\ & \left(\varepsilon^2 NQ_m^T(\varepsilon) f_0(\varepsilon) \right) + \frac{2mK_B T_g}{Me} \left(\varepsilon^2 NQ_m^T(\varepsilon) \frac{df_0(\varepsilon)}{d\varepsilon} \right) + \\ & \sum_J (\varepsilon + \varepsilon_J) f_0(\varepsilon + \varepsilon_J) NQ_J(\varepsilon + \varepsilon_J) \\ & - \varepsilon f_0(\varepsilon) N_J \sum_J Q_J(\varepsilon) = 0 \end{aligned} \quad (1)$$

Equation (1) is the so-called homogeneous electron Boltzmann transport equation and the isotropic component of the electron velocity distribution function, $f_0(v)$, can be expressed in terms of the electron energy $\varepsilon=0.5mv^2$, being then termed isotropic EEDF depends on the electron energy. K_B is the Boltzmann constant, T_g is the gas temperature, m/M is the ratio of electronic to atomic mass, $Q_J(\varepsilon)$ is the cross-sections for transitions from the ground state to the various excited states J, and ε_J is the threshold energy of the Jth excitation process, and $Q_m^e(\varepsilon)$ is the total effective momentum transfer cross section defined as follows,

$$Q_m^T(\varepsilon) = Q_m(\varepsilon) + \sum_J Q_e(\varepsilon) + Q_i(\varepsilon) + Q_a(\varepsilon) \quad (2)$$

Where, $Q_m(\varepsilon)$, $Q_e(\varepsilon)$, $Q_i(\varepsilon)$, and $Q_a(\varepsilon)$ are the electron cross-sections of momentum transfer, excitation, ionization, and attachment, respectively.

The 4th term in Equation (1) expresses inelastic loss processes due to electrons of energy $(\varepsilon + \varepsilon_J)$ undergoing a collision in which they lose the energy ε_J and appear as electrons of energy ε . The last term expresses gain of energy by electrons due to the second kind collision.

Equation (1) applies to swarm of electron drifting through gas and mixtures under the influence of a uniform dc electric field E in V/cm.

B. Transport Parameters

The electron transport coefficient in given gases calculated using a two-term approximation of the Boltzmann equation is functions of the density reduced electric field strength E/N, the gas temperature, and electron collision cross-section sets. The EEDF plays an important role in the calculation of electron swarm parameters.

The EEDF can be normalized by Colonna and D'Angola, 2016,

$$\int_0^\infty f(\varepsilon) \sqrt{\varepsilon} d\varepsilon = 1 \quad (3)$$

Taking into account, the normalization condition, $f_o(\varepsilon)$ and $f(\varepsilon)$ are linked to one another as follows,

$$f(\varepsilon) = \frac{1}{n_e} \frac{4\pi}{m} \sqrt{\frac{2}{m}} f_o(\varepsilon) \quad (4)$$

With such normalization, the Maxwellian electron distribution function at temperature T_g writes (Jiang and Economou, 1993),

$$f(\varepsilon) = \frac{2}{\sqrt{\pi}} (K_B T_g)^{-3/2} \exp\left(-\frac{\varepsilon}{K_B T_g}\right) \quad (5)$$

In terms of the EEDF, $f(\varepsilon)$, the mean electron energy is expressed as,

$$\langle \varepsilon \rangle = \int_0^\infty \varepsilon^{3/2} f(\varepsilon) d\varepsilon \quad (6)$$

With $\langle \varepsilon \rangle = \frac{3}{2} K_B T_g$, in the case of a Maxwellian, while the electron mobility $\mu_e = \sigma_e / en_e$, where σ_e the electron conductivity (Lee and More, 1984), is given by the equation,

$$\mu_e = -\frac{2}{3} \frac{e}{m} \int_0^\infty \frac{\varepsilon^{3/2}}{v_m^e(\varepsilon)} \frac{df}{d\varepsilon} d\varepsilon \quad (7)$$

Where, v_m^e is collision frequency, making the substitution $v_m^e = n_e Q_m^T$, we obtain the following expression for the reduced mobility,

$$N\mu_e = -\frac{1}{3}\sqrt{\frac{2e}{m}}\int_0^\infty \frac{\varepsilon}{Q_m^T(\varepsilon)} \frac{\partial f(\varepsilon)}{\partial \varepsilon} d\varepsilon \quad (8)$$

The electron drift velocity, diffusion coefficient, and characteristic energy are given as Smith and Thomson, 1978,

$$v_d = -\frac{\bar{E}}{3}\sqrt{\frac{2e}{m}}\int_0^\infty \frac{\varepsilon}{NQ_m^T(\varepsilon)} \frac{\partial f(\varepsilon)}{\partial \varepsilon} d\varepsilon \quad (9)$$

The transverse diffusion coefficient D_T is given by,

$$D_T = \frac{1}{3}\sqrt{\frac{2e}{m}}\int_0^\infty \frac{\varepsilon}{NQ_m^T(\varepsilon)} f(\varepsilon) d\varepsilon \quad (10)$$

$$\varepsilon_k = \frac{eD_T}{\mu_e} \quad (11)$$

From the computed drift velocity v_d , the ionization and attachment coefficients are obtained as Do, 2016,

$$\frac{\alpha}{N} = \frac{1}{v_d}\sqrt{\frac{2e}{m}}\int_i^\infty Q_i(\varepsilon)f(\varepsilon)\varepsilon^{1/2}d\varepsilon \quad (12)$$

$$\frac{\eta}{N} = \frac{1}{v_d}\sqrt{\frac{2e}{m}}\int_a^\infty Q_a(\varepsilon)f(\varepsilon)\varepsilon^{1/2}d\varepsilon \quad (13)$$

Where, $Q_i(\varepsilon)$ and $Q_a(\varepsilon)$ are ionization and attachment cross-section, here, i and a are the ionization and attachment threshold energy. The reduced limit electric field strength $(E/N)_{\text{limit}}$ is calculated when the formation and loss electrons reach a balance, this means that the effective ionization equal to zero $(\alpha-\eta)/N=0$ (Li, et al., 2012).

III. COLLISION CROSS-SECTION

The EEDF and values of transport coefficients in tetrafluoromethane (CF_4) gas calculated from the sets cross-section (elastic and inelastic) reported by Kurihara, et al., 2000. This set includes 16 collision processes: One momentum transfer cross-section (Q_m), three vibration excitations (Q_{v1} , Q_{v2} , and Q_{v3}) with threshold energy 0.0108, 0168, and 0.077 eV, respectively, and one electronic excitation (Q_{ex}) cross-section with threshold energy of 7.54 eV, one attachment cross-section (Q_a) with threshold energy 6.4 eV, seven dissociation ionization cross-sections (Q_{i1} - Q_{i7}) with threshold energy 16, 21, 26, 24, 34, 41, and 42 eV, respectively, and three neutral dissociation cross-sections (Q_{d1} - Q_{d3}) with threshold energy 12, 17, and 18 eV, respectively.

IV. RESULTS AND DISCUSSION

The EEDF and electron swarm parameters in pure tetrafluoromethane (CF_4) are calculated using the two-term approximation of Boltzmann equation in dc uniform fields in the range 0.1–1000 Td, at 1 Torr and 300 K.

Tetrafluoromethane gas has the momentum transfer cross-section which is greater than inelastic cross-section, this is a necessary condition for the two-term approximation solution of the Boltzmann equation to be valid (Smith and Thomson, 1978). The EEDF is one of the most important parameters for gas discharge phenomena and used for calculating electron swarm parameters. The E/N values were chosen to yield mean electron energies in the range of 0.045–14.3 eV. The EEDF for a dc field with various E/N values is given in Fig. 1 (where, E is the electric field and N is the gas density).

The influence of the EEDF versus the mean electron energy for different ratios of electric field strength (E/N) is shown in Fig. 1. The EEDF is strongly affected by changing parameter E/N, the higher values of E/N lead to the extension of the EEDF curves toward higher energy tail. The distribution function is normalized by Equation (3), the outcome of EEDF curves is due to constant electron density $n_e=1\times 10^{16} \text{ cm}^{-3}$. For the lowest $E/N \leq 1 \text{ Td}$, where $1 \text{ Td}=10^{-17} \text{ V.cm}^2$. The electron energy is thermal and the ionization degrees $(n_e/N) > (Q/10^{-13}) \varepsilon^2$ (where, ε is energy in eV and Q is an elastic and inelastic cross-section in the unit of cm^2) of the order of 10^{-3} – 10^{-4} can be sufficient to give Maxwellian characteristics of the EEDF as indicated by the straight line with a slope of $(-1/K_B T_e)$. In this case, the ionization degree is very small and the tetrafluoromethane (CF_4) atoms are in the ground state. However, in the range $E/N > 1 \text{ Td}$, the distribution is non-Maxwellian, the tail of the EEDF falls off rapidly as high-energy electrons are depleted due to different inelastic collisions. This behavior can be attributed to the electric field that heats the electrons and the energy of cold electrons increases. Subsequently, the mean electron energy depends on the electric field strength E/N.

Using EEDF, the electron swarm parameters have been calculated. Fig. 2 shows our calculated results for electron drift velocity as a function of E/N in comparison with the corresponding results provided by previous theoretical and experimental literatures.

The agreement between the present calculation and theoretical values (Bordage and Segure, 1999; Wu, et al., 2006; Duzkaya and Tezcan, 2019) and experimental values

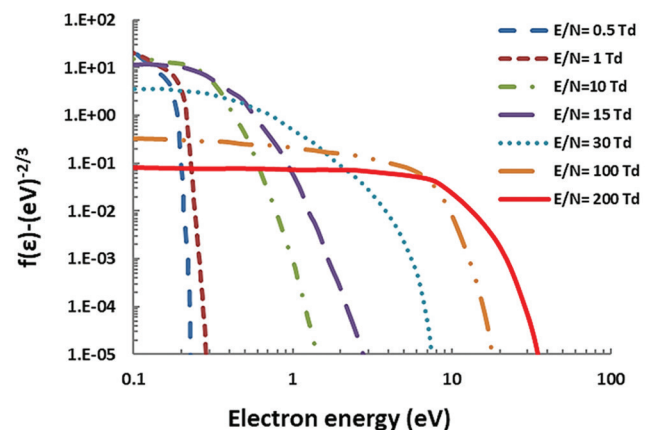


Fig. 1. Electron energy distribution functions in pure CF_4 for various E/N values.

(Hunter, et al., 1988; Nakamura, 1996; Xiao and Deng, 2013; Duzkaya and Tezcan, 2019) is excellent. The theoretical results of Sang-Nam, 2011, in range $E/N > 4$ Td are greater than present results and theoretical results of Michele, 2018, in the range $E/N \geq 50$ Td greater than present calculation. The drift velocity in pure CF₄ showed negative differential conductivity (NDC) over the range from 15 to 60 Td, the decrease in the drift velocity was not smooth but contained a small hump. A combination of the elastic and inelastic cross-sections can occur in a tetrafluoromethane (CF₄) molecules, at low electric field strength $E/N \leq 20$ Td the drift velocity rapidly increases, and at the region of higher E/N values ($20 \leq E/N \leq 100$ Td), the drift velocity will decrease with increasing E/N values. NDC will occur over a range of electric field strength E/N values in CF₄ when the inelastic cross-section has a large resonant type with respect to energies, and momentum transfer cross-section rapidly increases with electron energy.

Fig. 3 shows mean electron energy which increases with increasing E/N values, it is seen that the present calculation

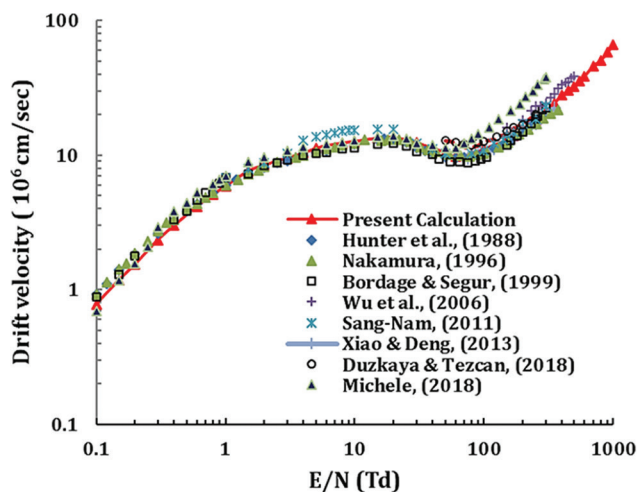


Fig. 2. Electrons drift velocity in pure CF₄.

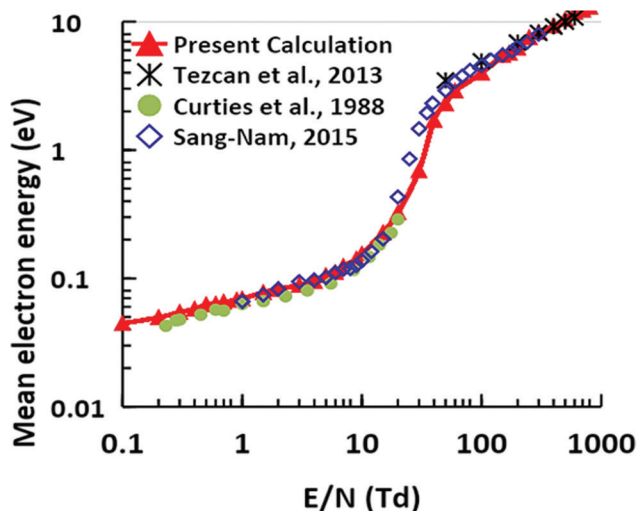


Fig. 3. Electron means energy in pure CF₄.

agree well with the experimental values of Curtis, et al. 1998, and theoretical values of Kurihara, et al., 2000; Tezcan, et al., 2013; Sang-Nam, 2015.

The calculated values of the characteristic energy of electrons as a function of E/N in pure CF₄ are shown in Fig. 4.

The calculated values compared with experimental values of Curtis, et al., 1988, and theoretical values of Mašek, et al., 1987; Stefanov and Pirgov, 1993; Bordage and Segur, 1999, a very good agreement was obtained, whereas the experimental values of Christophorou and Olthoff, 2004, are lower than calculated values over the E/N range between 2 and 300 Td. The characteristic energy increases with the increase of E/N during the inelastic collision process ($8 \text{ Td} \leq E/N \leq 80 \text{ Td}$) and then starts to be approximately constant at $E/N \geq 80$ Td, where above this value, the ionization process occurs for CF₄.

Fig. 5 shows density normalized electron mobility μN , which decreases with increasing E/N value, it is seen that the present calculation agree well with the experimental values of Hunter, et al., 1988.

The product of the longitudinal diffusion coefficient and the number density $ND_L(E/N)$ is shown in Fig. 6. The results

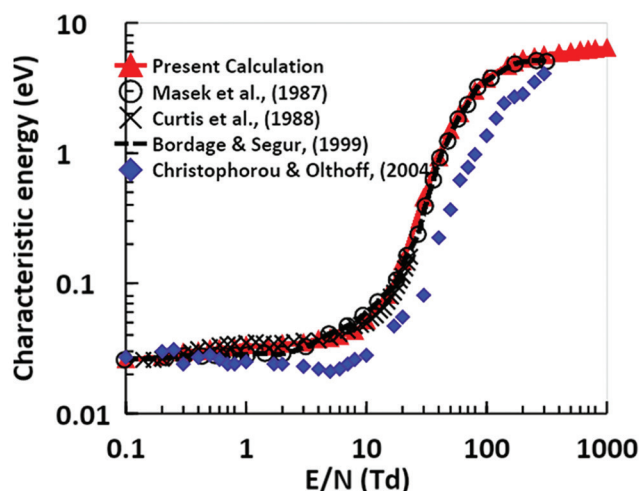


Fig. 4. Characteristic energy in pure CF₄.

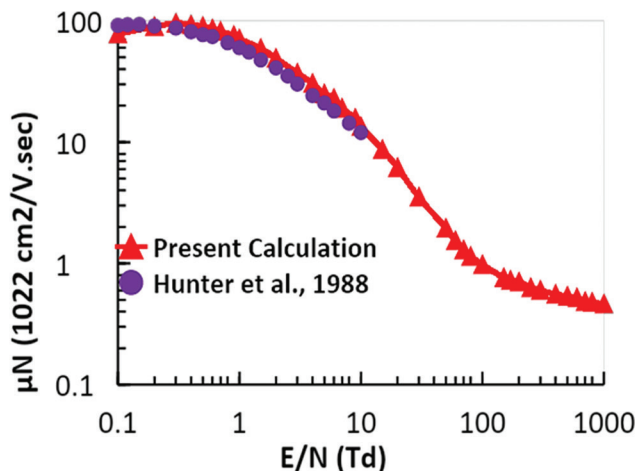


Fig. 5. Density normalized electron mobility μN in pure CF₄.

compared with the experimental values of Nakamura, 1996; Christophorou and Olthoff, 2004, and theoretical values of Kurihara, et al., 2000; Wu, et al., 2006.

The agreement is excellent over much of the range of E/N , with discrepancies in magnitude up to 10% in the region between 150 and 500 Td between present calculation and experimental values of Xiao and Deng, 2013. The density normalized ionization coefficient α/N has been calculated for the range $80 \leq E/N \leq 1000$ Td using Equation (12) and the results are shown in Fig. 7.

A good agreement has been obtained with the theoretical values of Mašek, et al., 1987; Bordage et al., 1999; San-Nam, 2012, and experimental values of Hunter, et al., 1988, for the whole range of E/N . Fig. 8 shows the density normalized attachment coefficient η/N as a function of E/N . Good agreement has been obtained with theoretical values of Bordage and Segur, 1999, and experimental values of Hunter, et al., 1988. The agreement between present data and theoretical values of San-Nam, 2012, reasonably good

over the entire range of E/N , with the largest discrepancies occurring between 120 and 300 Td.

The reduced limiting electric field strength $(E/N)_{\text{limit}}$ is an important quantity calculated from ionization and attachment coefficients. It is the value of the density reduced electric field at which $(\alpha-\eta)/N=0$. This value should also be equal to the breakdown voltage of CF_4 as calculated under the uniform field, this parameter is important for some high-voltage applications. The density normalized effective ionization coefficients $(\alpha-\eta)/N=0$ in pure CF_4 are shown in Fig. 9.

The change in the attachment coefficient is more obvious than the change in the ionization coefficient, so the change of the effective ionization coefficient is more depending on the change in the attachment coefficient. The present limiting electric field strength $(E/N)_{\text{limit}}$ of pure CF_4 is evaluated as 146 Td, which is comparable with experimental values reported by Christophorou and Olthoff, 2004, and theoretical values of Liu and Xiao, 2007; Duzkaya and Tezcan, 2019, where the estimated values of $(E/N)_{\text{limit}}$ are 146, 144, and 145 Td, respectively.

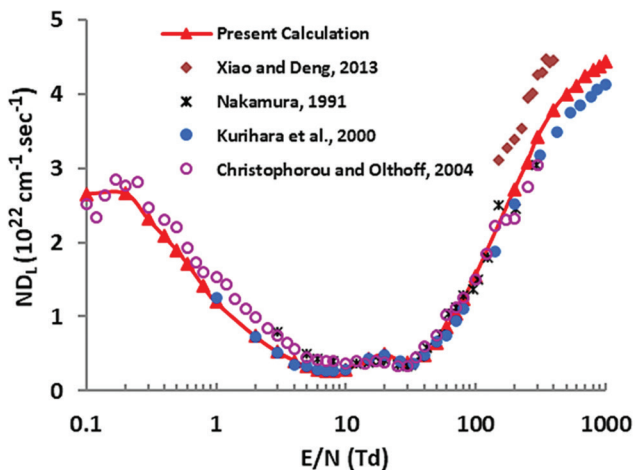


Fig. 6. The product of the longitudinal diffusion coefficient and the gas number density in pure CF_4 .

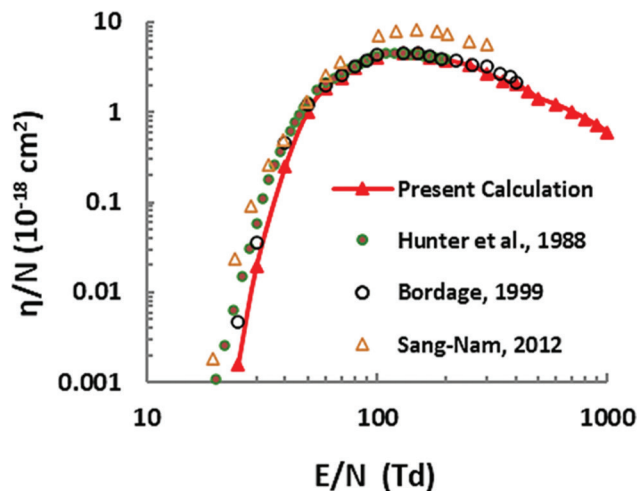


Fig. 8. Attachment coefficient in pure CF_4 .

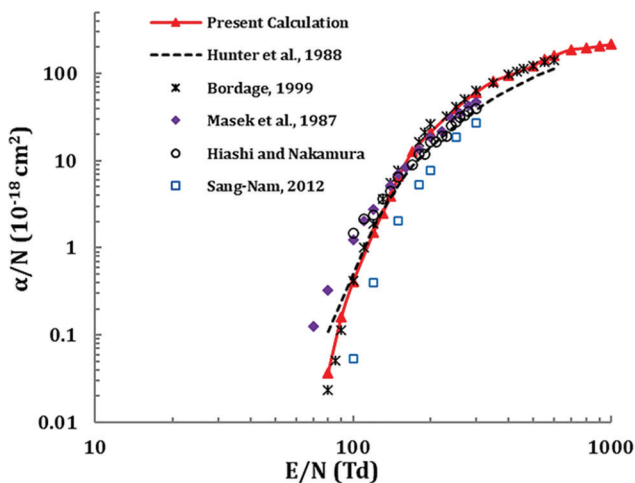


Fig. 7. Ionization coefficient in pure CF_4 .

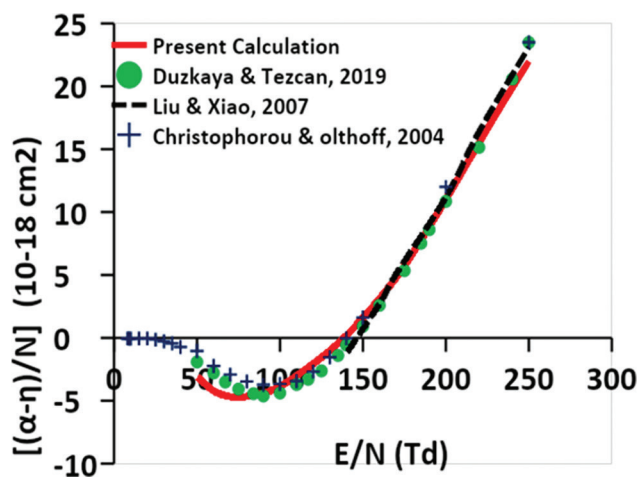


Fig. 9. Effective ionization coefficient $(\alpha-\eta)/N$ in pure CF_4 .

V. CONCLUSION

Tetrafluoromethane (CF_4) is an electronegative and potent greenhouse gas. In this study, the EEDF, electron swarm parameters, and density normalized effective ionization coefficient of tetrafluoromethane (CF_4) are calculated using the two-term approximation solution of Boltzmann equation method for the range reduced electrical field strength varying from 0.1 Td to 1000 Td at temperature 300K. The present results based on electron collision cross-section sets for CF_4 molecules. The EEDF is strongly affected by changing the parameter E/N, we have shown that the EEDF in pure CF_4 shifts toward increasing of mean electron energy according to the increasing of E/N. The calculated swarm parameters (drift velocity, characteristic energy, mean electron energy, diffusion coefficient, electron mobility, ionization, and attachment coefficient) agree well with experimental and theoretical values. The reduced limiting electric field strength $(E/N)_{\text{limit}}$ at which the ionization and attachment rate are exactly balanced as a function of E/N, has been calculated from the effective ionization curves.

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