

RESEARCH PAPER

Boltzmann equation studies on electron swarm parameters for oxygen plasma by using electron collision cross – sections

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ABSTRACT:

The Boltzmann transport equation has been solved using a two-term approximation method in pure electronegative gas oxygen to evaluate the electron energy distribution function (EEDF) and electron transport parameters for a wide range of E/N varying from 0.1 to 1000 Td (1 Td=10⁻¹⁷ V.cm²). These parameters, are “electron drift velocity, mean electron energy, characteristic energy, diffusion coefficients, electron mobility, attachment and ionization coefficients, effective *ionization coefficient and critical reduced electric field strength (E/N)_{crit}*”. The dependence of second kind collision (super-elastic collision) and electron energy distribution function on E/N are explained (where E is electric field and N is neutral number density). The present calculated results are in good agreements as compared, with the previous experimental and theoretical results. A group of electron/molecule collision (elastic and inelastic) cross-sections are collected for oxygen gas to evaluate transport parameters over the entire E/N range. In addition, the energy lost by different types of electron/molecule collision processes are computed as a function of E/N.

KEY WORDS: Boltzmann equation, Electron energy distribution function (EEDF), Electron transport parameters, Critical field strength, Electric discharge.

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1. INTRODUCTION

Oxygen molecule (O₂) is a strongly electro negative gas, is an electron attaching gas (O₂ + e → O + O⁻) and it decreases with the electrical discharge, which gives oxygen an excellent dielectric strength (McNevin, 1990). Oxygen is the one of the main combinations of the earth's atmosphere (20.8%) and is the third most numerous elements in the Universe after hydrogen and helium gases. Oxygen is an environmentally clean, nontoxic, nonflammable, colorless, orderless diatomic gas and non-reactive but an oxidizer, and it can be toxic at elevated partial pressure (more than 160mmHg). It has a high critical temperature (-154.6K) and has high critical pressure (49.8 atm).

At low temperature plasma oxygen molecule with noble gases and molecules are used in application of various fields such as: material processing properties which used as an arc quenching medium. (Harthney et al., 1989) biomedical purposes (Graves, 2014) environmental /energy application (Tatarova, et al., 2014) in material processing, such as photoresist aching, surface modification, chemical vapor deposition (VCD) and oxidation. Plasma discharges of Cl₂ and O₂ mixtures were used in application thin film etching silicon (McNevin, 1990) and deposition in microelectronic device fabrication (Thorsteinsson. and Gudmundsson, 2010). In addition, industrial oxygen mixed with pure CF₄ plasma to control the production of fluorocarbon (CF) molecules which were

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generally used for etching of Si and SiO₂ (Lu *et al.*, 2012). Because of its industrial importance and for understanding plasma phenomena a set of electron/molecule cross-sections for pure oxygen evaluated by (Phelps, 1985; Jeon, 2003). The electron swarm parameters were studied experimentally and theoretically by variety of investigators as a function of electric field strength E/N. For example (Settaouti, 2007) calculated electron swarm parameters over the range 100 Td -1000 Td, (1Td=10⁻¹⁷ V.cm²). The attachment and ionization coefficients are experimentally measured by (Jeon *et al.*, 1997). At low electric field strength electron swarm parameters in N₂, O₂ and H₂ are theoretically calculated by (Ridenti *et al.*, 2015) using homogenous Boltzmann equation. For the purpose of abbreviation, previous literatures of electron swarm parameters in pure oxygen are collected in table 1.

The Monte-Carlo simulation method used by (Leyla *et al.*, 2011) to evaluate the mean electron energy, ionization and attachment coefficients as functions of time for an electrical discharge in pure oxygen molecule and (Settaouti, 2018) studied the point plane corona discharge by using an equivalent method. The electron energy distribution function (EEDF) plays a crucial role in calculating electron swarm parameters and physical properties of plasma, (Nighan, 1970; Engelhardt and Phelps, 1963; Jiang and Economou, 1993) theoretically calculated the electron swarm parameters using (EEDF) by two-term approximation solution of Boltzmann equation under dc field. Electron swarm parameters in oxygen were also calculated in binary gas mixtures O₂-Ar (Jeon and Nakamura, 1998), SF₆-O₂ (Hernandez-Avila and Urquijo, 2006; Linsheng *et al.*, 2014), Cl₂-O₂ (Tuan, 2014), O₂-CO (Price and Moruzzi, 1973), N₂-O₂ (Dujko *et al.*, 2011; Pancheshnyi, 2013; Guerra *et al.*, 2019), TMS-O₂ (Hien, *et al.*, 2012), TEOS-O₂ (Yoshida *et al.*, 1996; Tuan and Jeon, 2012), CO₂-O₂ (Yousfi *et al.*, 2009) and TRIES-O₂ gas mixtures (Tuoi *et al.*, 2018). The ternary mixtures of CF₄/Ar/O₂ were analyzed by (Nikitovic *et al.*, 2010; Nikitovic *et al.*, 2011) Furthermore, ternary mixtures of C₅F₁₀O/CO₂/O₂ was used in gas-insulated high-voltage switchgear (GIS) (Bousoltane *et al.*, 2018) and C₄F₇N or C₅F₁₀O

mixtures with CO₂ and O₂ were used as insulating and switching media in the case of high and medium voltage (Eguz *et al.*, 2019).

Electrical breakdown voltage and critical field strengths (E/N)_{crit.} were calculated at condition that reduced ionization coefficient (α/N) and reduced attachment coefficient (η/N) are in equilibrium, therefore the effective ionization coefficient equal to zero ($\bar{\alpha} = (\alpha/N) - (\eta/N) = 0$), this method was used by (Láska *et al.*, 1984; Brand and Kopainsky, 1979; Itoh *et al.*, 1980; Zhao *et al.*, 2017) to calculate electrical breakdown voltage using two-term approximation solution of Boltzmann equation. Critical field strength have been studied by both theoretical calculations (Tanaka, 2004; Rong *et al.*, 2014; Zhao and Lin, 2016; Zhao *et al.*, 2017) and experimentally to find new gases with high dielectric field strength which used in electrical insulator, for example, experimentally (Xiao *et al.*, 2018) reported that small addition of oxygen O₂ reduced breakdown voltage of the c-C₄F₈/N₂ mixed gas. (Beatty, *et al.* 1979 and Dutton, 1975) published a large data on the electron swarm parameters in number gases over a wide range of electric field strength E/N. We previously reported a detailed explanation to calculate electron swarm parameters by using two-term approximation method of the Boltzmann equation (Othman, *et al.* 2018; Othman, *et al.* 2019a).

In the present work, the electron swarm parameters and critical field strength (E/N)_{crit.} for a wide range of E/N varying from 0.1Td to 1000 Td in pure oxygen gas are calculated using a two-term approximation of the Boltzmann equation. Since 1970, several numerical techniques have used to calculate electron, energy distribution, function and electron swarm parameters. BOLTZ code (Thomson and Smith, 1976), NOMAD code (Rockwood and Greene, 1980), ELENDIF code (Morgan and Penetrante, 1990), METHES code (Rabie and Franck, 2016), and LoKI-B (Tejero-del-Caz, *et al.*, 2019). NOMAD and ELENDIF code are containing electron attachment coefficient which are not considered by BOLTZ code. In the present study one chooses the NOMAD code, because it's more limited by available of electron energy collision cross-sections as compared to ELENDIF code.

Table 1. Summary of previous theoretical work (Boltzmann equation & Monte-Carlo method)

<i>Investigator</i>	<i>E/N range (Td)</i>	<i>v_d cm/s</i>	<i><u> eV</i>	<i>u_k eV</i>	<i>η/N cm²</i>	<i>α/N cm²</i>	<i>f(x) eV^{-3/2}</i>
Hake and Phelps, 1967	0.01-150	x		x	x	x	x
Myers, 1969	0.003-200		x	x			x
Wanger, 1971	90-50				x	x	
Nelson & Davis, 1972	≥1.3	x					
Lucas et al., 1973	15-152	x	x		x	x	x
Crompton & Elford, 1973	0.8-12	x					
Mašek, 1975	1-140	x	x	x		x	x
Mašek et al., 1977a	1-200	x	x	x			x
Mašek et al., 1977b	10-200				x		x
Taniguchi et al., 1978	1-30				x		
Reid & Crompton, 1980	0.14-1.4	x					
Roznerski and Leja, 1984	50-250	x					
Al-Amin et al., 1985	14.1-5650	x	x	x		x	
Gousset et al., 1991	0-130	x	x	x		x	x
Liu and Govinda Raju, 1992	20-5000	x	x	x	x	x	x
Liu and Govinda Raju, 1993	20-2000	x	x			x	
Jeon, 2003	1-1000	x		x	x		
Rabie & Frank, 20016	≥200	x	x				
Alves et al., 2016	10 ⁻³ -1000			x	x	x	
Mašek, 1975	1-140	x	x	x		x	x

2. Theory

2.1 The Boltzmann Equation

The electron energy distribution function (EEDF) derived from the solution of the Boltzmann equation. This parameter used to

$$\frac{\partial f(v)}{\partial t} + V \cdot \nabla f(v) - \frac{e}{m} \bar{E} \cdot \nabla_v f(v) = \left(\frac{\partial f}{\partial t} \right)_{coll} \quad (1)$$

where $f(v)$ is the electron energy distribution function, V is the velocity, m is the electron mass, \bar{E} is the dc electric field and ∇_v is the velocity-gradient operator. The right part of the equation denotes the rate of change in the electron distribution due to elastic and inelastic collisions. To solve the Boltzmann transport equation the electron distribution function is expanded in two-terms of Legendre polynomials, $f(\vec{v}) = f_o(v) + f_1(v) \cos \theta$, where $f_o(v)$ is the isotropic part and $f_1(v)$ is anisotropic part of the distribution function where $f_1(v) \ll f_o(v)$. The two-term approximation is used to deduce the swarm equations by filling them into the Boltzmann equation. (Govinda-Raju, 2006 and 2012) collected a large literatures on the solution of Boltzmann equation by the numerical method.

),

$$f(u) = 2 * \left(\frac{u}{\pi (K_B * T_e)^3} \right)^{1/2} \exp\left(\frac{-u}{K_B T_e} \right) \quad (2)$$

where T_e is electron temperature, this leads to an electron mean energy of,

$$\langle u \rangle = \int_0^{\infty} u f_0(u) du = \frac{3}{2} T_e \quad (3)$$

where T_e is expressed in electron volts.

However, in many cases, deviations occur and non-thermal plasma often possess, the distribution follows a non-Maxwellian shape (Hagelaar and Pitchford, 2005). In this case, the electron energy distribution function (EEDF) derived analytically

$$v_d = -\frac{1}{3} \sqrt{\frac{2e}{m}} \frac{\bar{E}}{N} \int_0^{\infty} \frac{u}{Q_m^T(u)} \frac{\partial f_0(u)}{\partial u} du \quad (4)$$

calculate reaction rates and electron swarm parameters in pure gases/mixtures. The general form of Boltzmann equation written as follows, (Morgan and Penetrante, 1990; Govinda Raju, 2017).

2.2. The Transport Parameters

The electron transport coefficient in gases is calculated by using a two-term approximation solution of the Boltzmann equation are functions of E/N the gas temperature (T_g) and electron/molecule (atom) cross-sections. The relation between E/N and E/p is $E/N [\text{Vcm}^2] = 1.036 \times 10^{-19} T_g [\text{K}] \cdot E/P [\text{Vcm}^{-1} \text{Torr}^{-1}]$, where T_g is gas temperature, for example at $T_g = 273 \text{K}$, $E/P = 1 \text{Vcm}^{-1} \text{Torr}$ then $E/N = 2.823 \text{Td}$, ($1 \text{Td} = 10^{-17} \text{Vcm}^2$). The electron energy distribution function is the important parameter used for calculating electron swarm parameters, for thermal equilibrium the electron distribution function is given by Maxwellian distribution as follows, (Fridman, 2008),

is the main coefficient to calculate the electron swarm parameters, that is. the electron drift velocity v_d and the transverse diffusion coefficient D_T as follows (Thomson and Smith, 1976; Al-Amin and Lucas, 1988),

$$D_T = \frac{1}{3N} \sqrt{\frac{2e}{m}} \int_0^\infty \frac{u}{Q_m^T(u)} f_0(u) du \quad (5)$$

(Sakai et al., 1977) measured longitudinal diffusion coefficient experimentally using time-of-flight method.

The electron mobility, $\mu_e = v_d \bar{E}$ and the mean electron energy $\langle u \rangle$, in terms of the electron

$$\mu_e = -\frac{1}{3N} \sqrt{\frac{2e}{m}} \int_0^\infty \frac{u}{Q_m^T(u)} \frac{\partial f_0(u)}{\partial u} du \quad (6)$$

$$\langle u \rangle = \frac{2}{3} \int_0^\infty u^{3/2} f_0(u) du \quad (7)$$

here, $Q_m^T(u) = Q_m(u) + \sum_j Q_k(u) + Q_i(u) + Q_a(u)$ represent the total effective momentum transfer cross section. Where $Q_m(u)$, $Q_k(u)$, $Q_i(u)$ and $Q_a(u)$ are momentum transfer, excitation

$$u_k = e \frac{D_T}{\mu_e} = e \frac{D_T}{v_d} \bar{E} \quad (8)$$

whereas for the thermal equilibrium,

$$u_k = \frac{2}{3} \langle u \rangle \quad (9)$$

where $u_k = \frac{3}{2} K_B T_g$, the following relation obtained,

$$\frac{D_T}{\mu_e} = \frac{K_B T_g}{e} \quad (10)$$

This is also known as Einstein's relation.

By using the drift velocity v_d equation, the reduced ionization and attachment coefficients are calculated as follows, (Lucas et al., 1973; Láska,

$$\frac{\alpha}{N} = \frac{1}{v_d} \left(\frac{2e}{m} \right)^{1/2} \int_{u_i}^\infty u Q_i(u) f_0(u) du \quad (11)$$

energy distribution function (EEDF) (Smith and Thomson, 1978; Smith and Thomson, 1978; Hagelaar and Pitchford, 2005; Ridenti and Amorim, 2012), are expressed as follows,

(vibration/ electronic), ionization, and attachment cross sections respectively.

The definition of characteristic energy u_k is given by combining equations (5) and (6), (Makabe and Petrovic, 2015) which yields:

et al. 1984; Loureiro and Amorim, 2016; Othman, et al., 2019b),

$$\frac{\eta}{N} = \frac{1}{v_d} \left(\frac{2e}{m} \right)^{1/2} \int_{u_a}^{\infty} u Q_a(u) f_0(u) du \quad (12)$$

Where, $Q_i(u)$, $Q_a(u)$ are ionization and attachment cross section, here, u_i is the ionization threshold energy for oxygen which is equal to 12.2 eV, and u_a is attachment threshold energy of 4.4 eV. The reduced critical electric field strength $(E/N)_{\text{crit}}$ is calculated when reduced

$$\bar{\alpha} = \frac{\alpha}{N} - \frac{\eta}{N} = \frac{\alpha - \eta}{N} = 0 \quad (13)$$

The rate constant of excitation for the j^{th} inelastic collision cross-sections is obtained from information of cross-sections and electron energy

$$R_{sj} = \left(\frac{2e}{m} \right)^{1/2} \int_0^{\infty} u N Q_{sj}(u) f(u) du \quad (14)$$

where Q_{sj} is electron cross-sections of excitation of level j in species, s .

$$P_j = \frac{u_j R_{sj}}{e E v_d} \quad (15)$$

Where u_j is the onset energy for the excitation. The neutral number density for pure gas calculated as follows,

$$N = \frac{\rho N_A}{M_w} \quad (16)$$

where ρ represents gas density, M_w is molecular weight and N_A is Avogadro number.

ionization coefficient (α/N) and the reduced attachment coefficient (η/N) are in balance, in this case, the effective ionization coefficient ($\bar{\alpha} = 0$) equal to zero (Láska et al, 1984; Li et al., 2012).

distribution function by the following formula (Nakamura and Lucas, 1978)

The electron energy loss P_j during inelastic collision process is calculated as follows,

3. The Cross Section

The electron/molecule collision cross-sections (Phelps, 1985) are necessary in order to calculate the EEDF and electron swarm parameters in oxygen gas. The oxygen molecule includes 14 sets of collisional cross-sections: one momentum transfer cross-sections (Q_m), eight vibrational excitation (Q_{v1} , Q_{v2} , Q_{v3} , Q_{v4} , Q_{v5} , Q_{v6} , Q_{v7} , Q_{v8}) with threshold energy 0.37, 0.56, 0.75, 0.93, 1.12, 1.3, 1.47 and 1.46 eV, respectively, three electronic excitation (Q_{ex1} , Q_{ex2} , Q_{ex3}) cross-sections with threshold energy of 4.4, 8.0 and 9.7 eV, respectively, and one attachment cross-sections (Q_a) with threshold energy 4.4 eV are taken from (Hake and Phelps, 1967), and one dissociation ionization cross-sections with threshold energy of 12.2 eV is taken from (Rapp and Englander-Golden, 1965).

4. Results and Discussion

There are several literatures published of the electron swarm parameters in pure oxygen gas, however it is impossible to display all the previous experimental and theoretical data. For comparison with the present results one shows only selected data with an emphasis on more recent results. The major objective of this research is the calculation of the reduced critical electric field strength and electron swarm parameters for pure oxygen in the range 0.1 Td to 1000 Td by using two term solution of Boltzmann equation.

Normalized electron energy distribution function (EEDF) for various reduced electric field strength E/N in pure oxygen molecule at temperature 300K and pressure 1 atm are shown in figure (1). When the electron energies ≤ 4.4 eV, the EEDF decreases as the reduced electric field strength E/N increases, the tail of the distribution is decreasing and there are only small number of electrons that have energies greater than the ionization potential. Moreover, due to inelastic collision there are few electrons with energies, greater than the excitation energy. At low electron energy the shape of the energy distribution function depends on the momentum transfer cross-sections, but for electron energies >4.4 eV, the EEDF increases with increasing E/N , then the electrons gain at higher kinetic energy, and the tail extends to energies above the ionization potential. As E/N increases degree of ionization increase and then increases the number of particles with energies higher than excitation, energy tends to spread energy due to

collisions, at high energies the distribution function influenced by electron collisions so the distribution approaching to straight line becomes more nearly of the Maxwellian form, with a slope of, $(-1/k_B T)$.

Figures (2a) and (2b) are shown the influence of second kind collisions (super-elastic collisions) from metastable electronic states on EEDF at electric field strength 0.5 Td and 10 Td with and without super-elastic collision respectively. Figure (2b) shows, that super-elastic collisions are not important at higher reduced electric field strength E/N .

The electron drift velocity as a function of E/N is shown in figure (3) which increases with increasing E/N values, the results are compared with the theoretical and experimental results are also illustrated in the same figure, a good agreement is obtained when compared with experimental results of (Roznerski and Leja, 1984; Al-Amin et al., 1985; Jeon et al., 1997). While the theoretical results calculated by (Liu and Govinda-Raju, 1992, Liu and Govinda-Raju, 1993; Jeon, 2003; Settaouti and Settaoutim 2007 and Tuoi and Tuan, 2018) are observed to be in good agreement over the common E/N range. Figure (4) illustrates the mean electron energy in range of 0.12 eV-19.39 eV for different values of E/N varying from the range 0.1 Td to 1000 Td. The variation of mean electron energy progresses exponential the electrons gain energy from the electric field, comparison has been made with the theoretical values of (Liu and Govinda Raju, 1992, 1993; Settaouti and Settaouti, 2005 and Dujko et al., 2011) a good fit has been observed. Figure (5) shows the calculated characteristic energy as a function of E/N , during inelastic collision process $E/N \geq 40$ Td, the ionization processes occur and then the characteristic energy starts to increase with increasing E/N . In comparison the present results are found to be in good fit with theoretical results of (Hake and Phelps, 1967; Jeon, 2003) and experimental results of (Al-Amin et al., 1985). Otherwise the experimental values of (Jeon and Nakamura, 1998) and theoretical values of (Toui and Tuan, 2018) are lower than the present results at the range $E/N \geq 1$ Td, the variation of present results gives rise by large momentum transfer cross-section.

Figure (6) is comparison between the present results for electron mobility with theoretical results of (Alves et al., 2016) at room

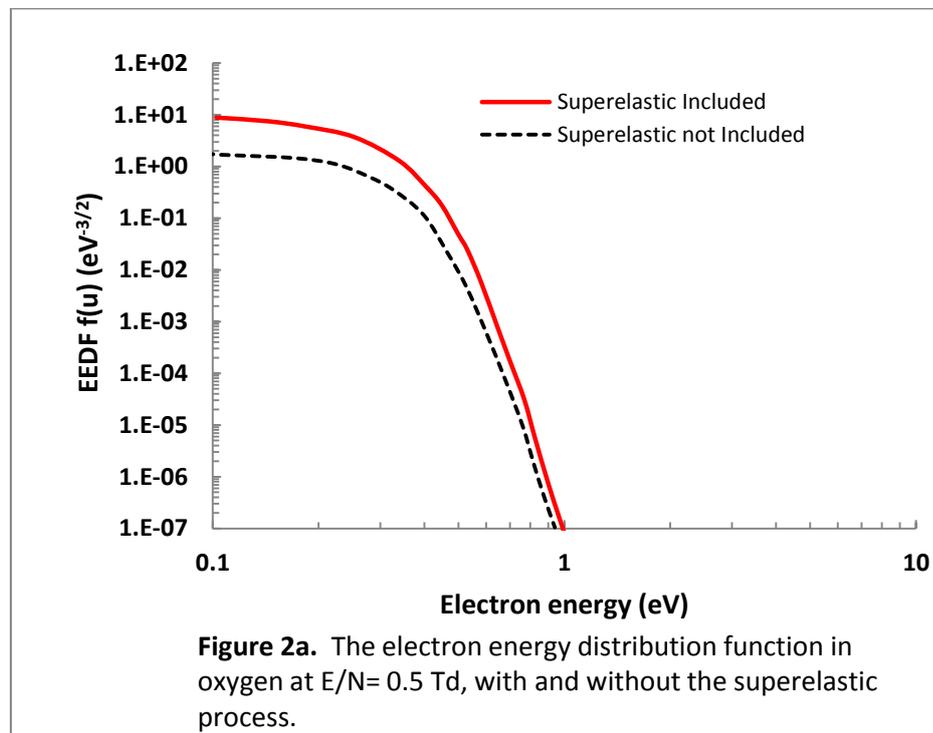
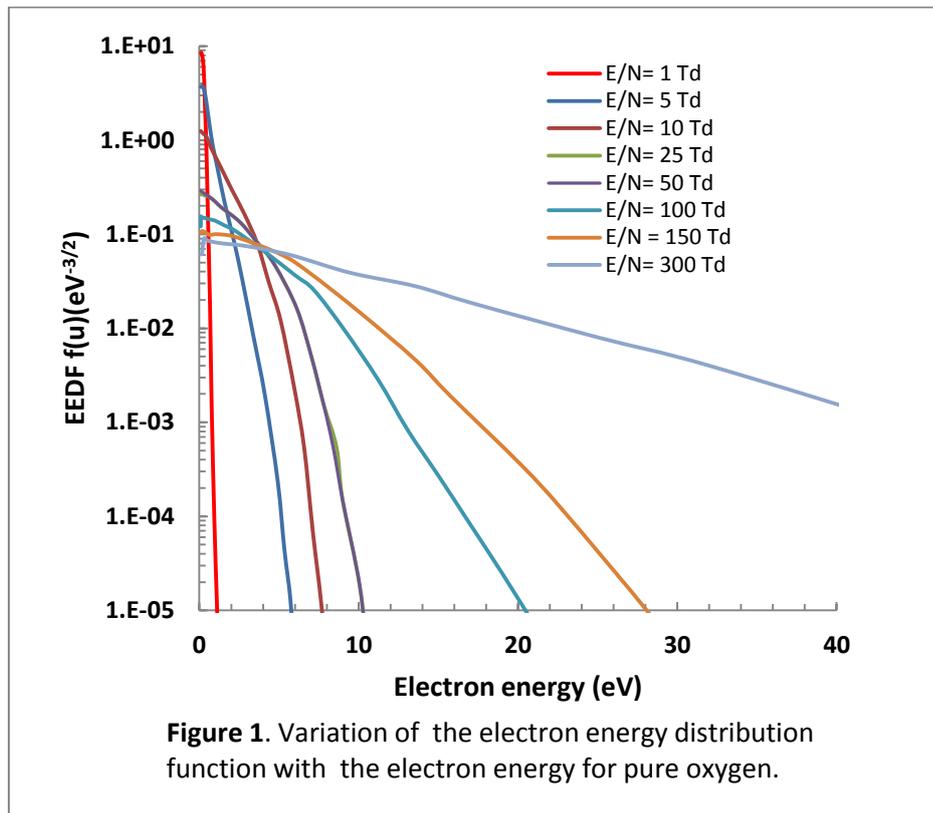
temperature, the present results agree well with theoretical results. The transverse diffusion coefficient, DN for electrons in pure oxygen, as a function of E/N varying from 0.1 Td to 1000 Td is illustrated in figure (7), together with theoretical multi term solution results obtained from (Dujko, et al., 2011) for comparison. It indicates the good agreement between present results and theoretical results of (Dujko, et al., 2011).

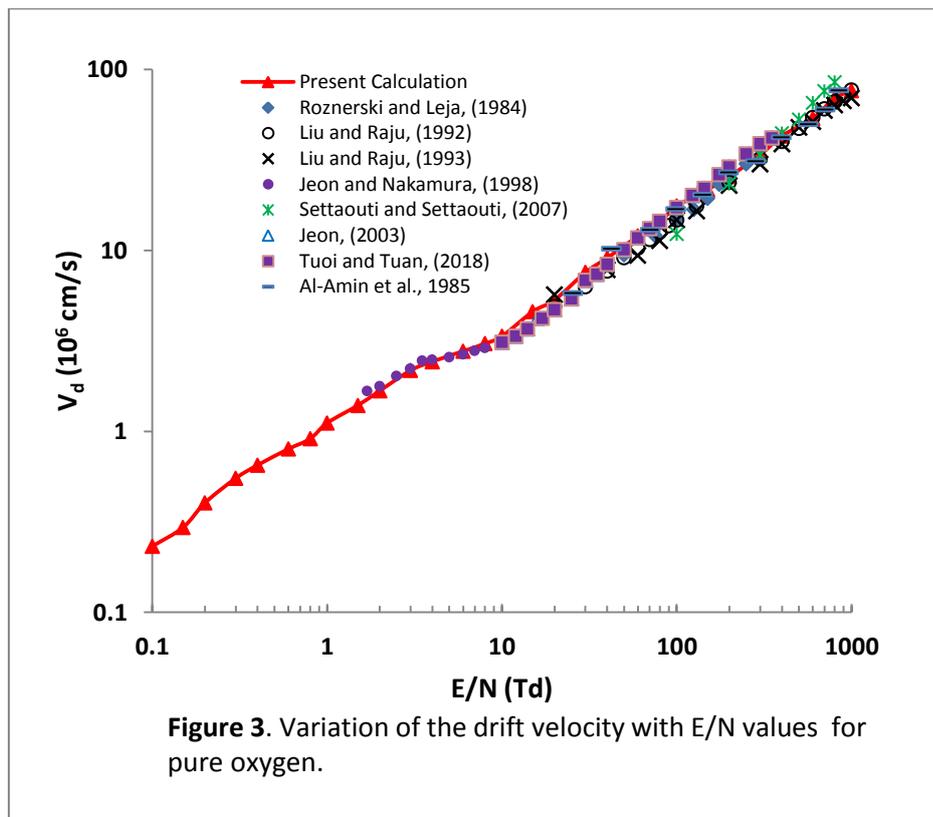
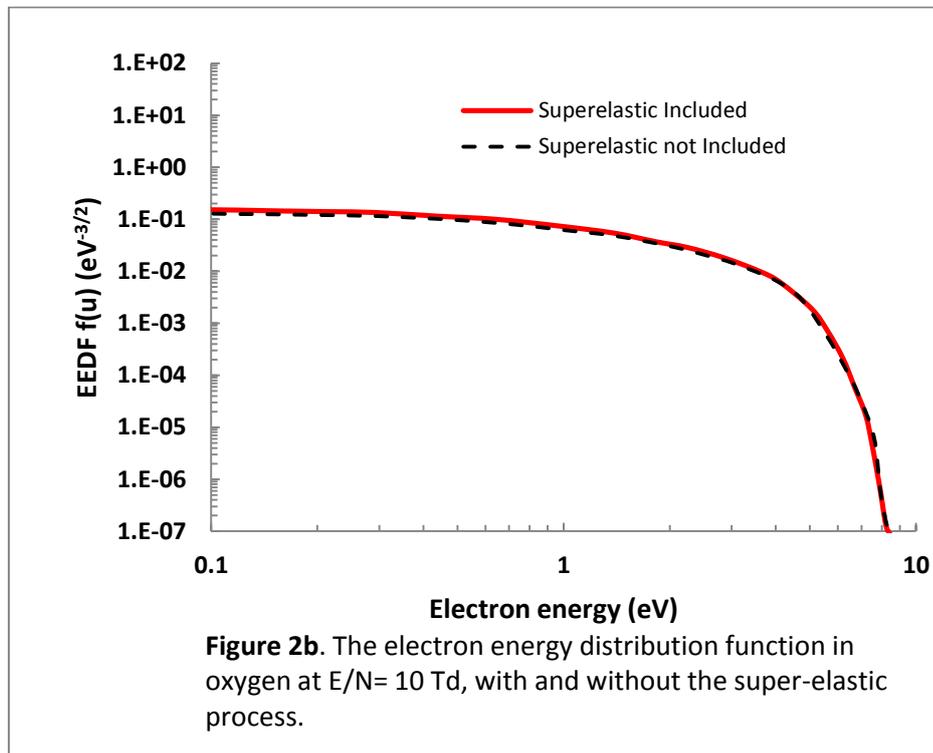
Figure (8) shows the Townsend, ionization coefficient α/N (also known as reduced ionization coefficient) is calculated for the range $40 \leq E/N \leq 1000$ Td. The ionization coefficient increases as E/N increases, good agreement has been obtained with the theoretical values of (Liu and Govinda Raju, 1993; Settaouti and Settaouti, 2007 and Tuoi and Tuan, 2018) and the experimental results of (Al-Amin et al., 1985 and Yoshida et al., 1996). 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 Townsend reduced, attachment coefficient, η/N (also known as attachment, reduced coefficient), for the pure oxygen molecule as a function of E/N is shown in figure (9). The present calculation is found to be in a good agreement with theoretical results of (Jeon, 2003; Hien et al., 2012 and Alves et al., 2016) and experimental results of (Huxley et al., 1959). An electron avalanche can occur when the effective ionization-coefficient $(\alpha-\eta)/N > 0$.

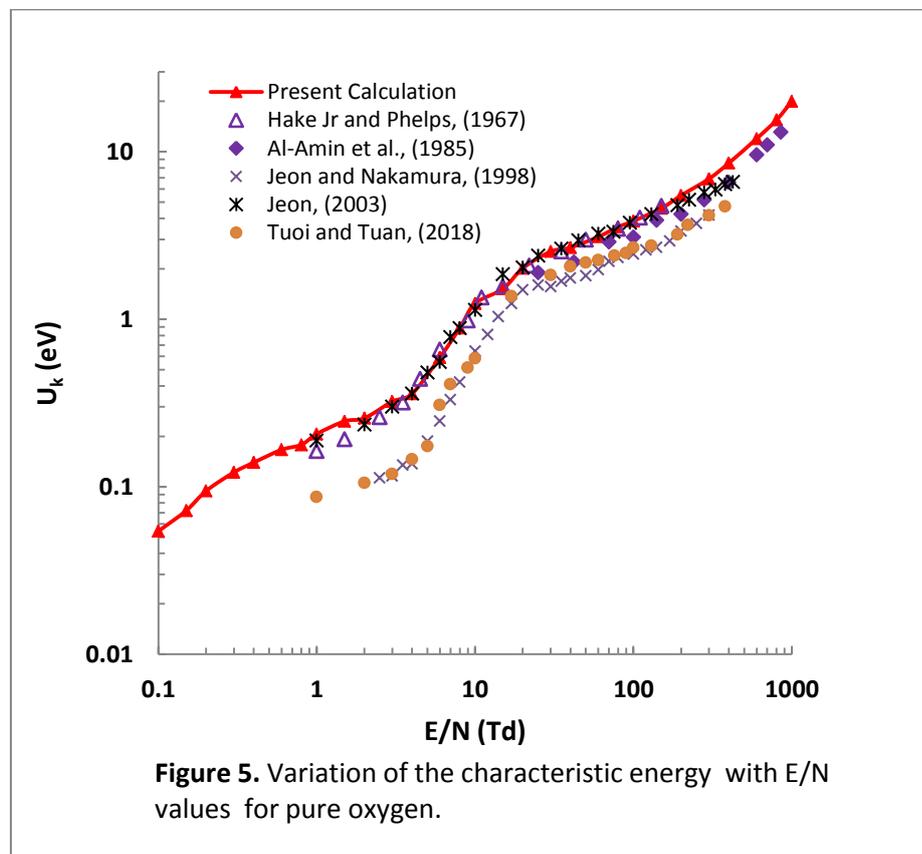
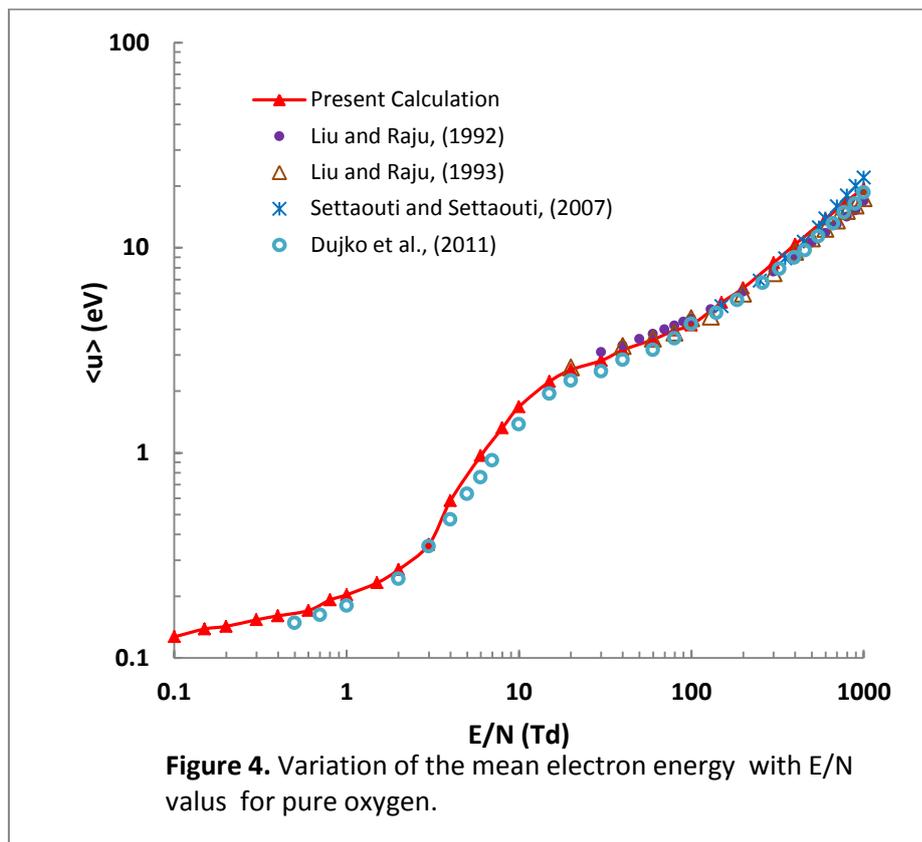
The reduced effective ionization coefficient $(\alpha-\eta)/N$ as a function of E/N in pure oxygen is shown in figure (10) which calculated from the results of α/N and η/N . The critical reduced electric field strength, $(E/N)_{\text{crit}}$ is

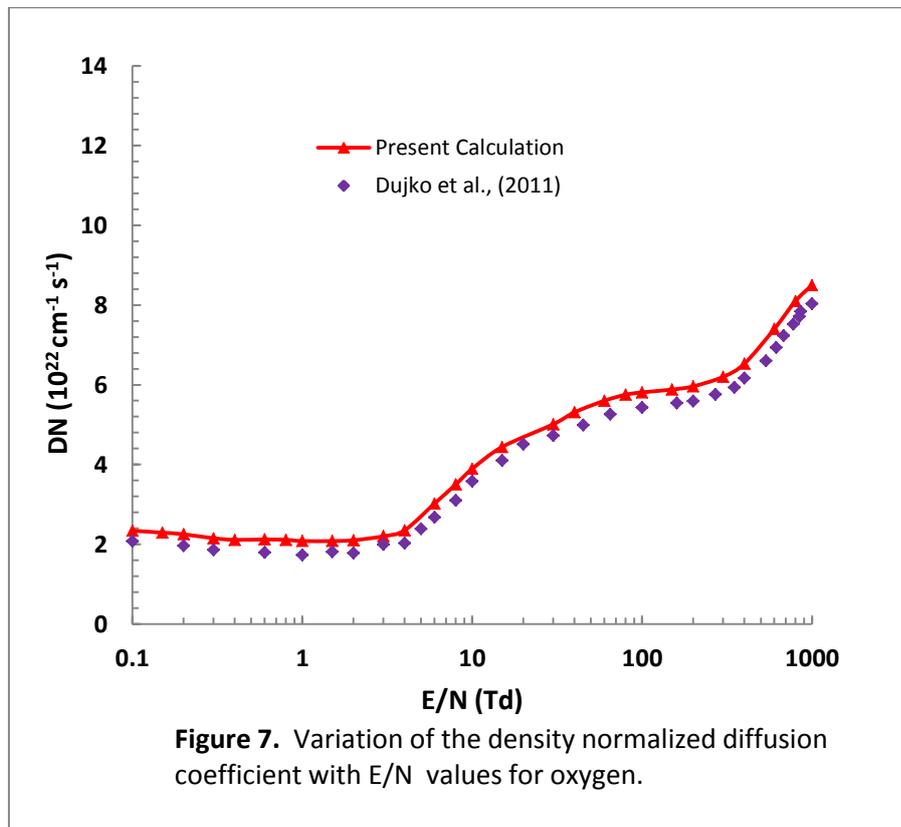
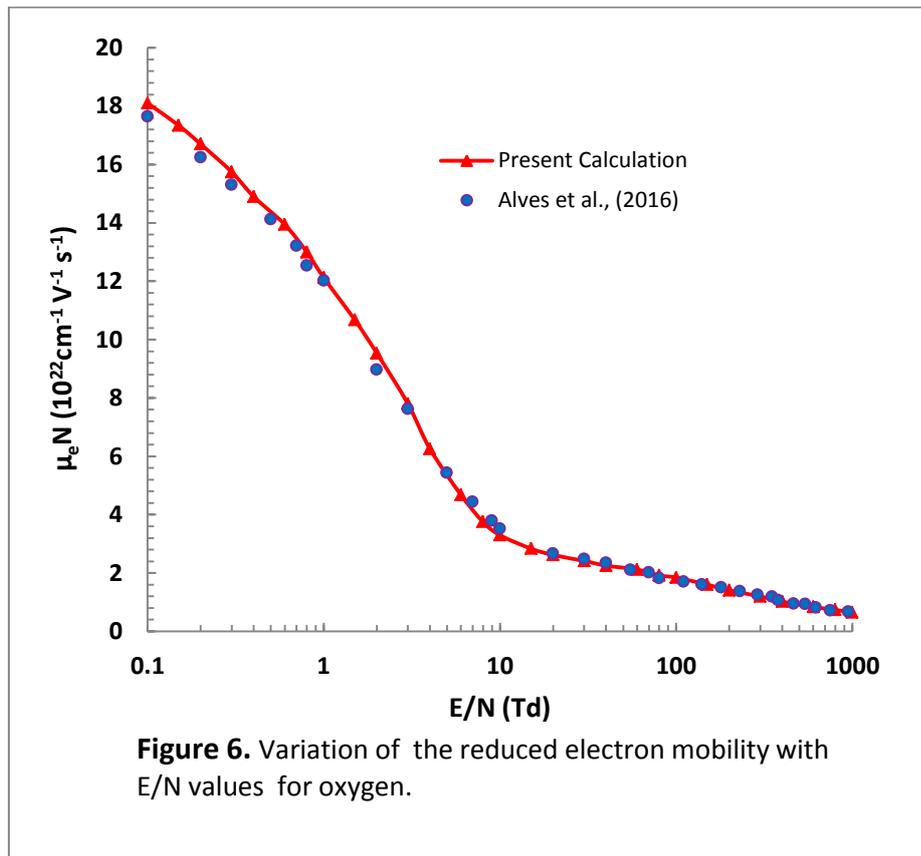
described, as the value of E/N at $(\alpha-\eta)/N = 0$, calculated from the results of the effective ionization coefficient $(\alpha-\eta)/N$ which describes variations of free electrons in oxygen.. Critical reduced electric field strength, $(E/N)_{\text{crit}}$ is important coefficient for the purpose of identification the insulation performance of electronegative gases. The $(E/N)_{\text{crit}}$ value is shown in figure (10), at which $\alpha/N = \eta/N$, in the present calculation $(E/N)_{\text{crit}}$ equal to (119 Td), in agreement for comparison with the results of (Laska et al., 1984), 110 Td, (Tuan, 2014), 118.5 Td and (Zhao et al., 2017), 120.5 Td. The reduced effective ionization coefficient is zero, since α/N are balanced with η/N , when $E/N < (E/N)_{\text{crit}}$. attachment processes dominants, in this case negative values for the effective ionization coefficient as E/N is decreased, on the other hand, for $E/N > (E/N)_{\text{crit}}$. the effective ionization coefficient increases with increasing E/N values where the ionization collisions dominants, in this case, the effect of the attachment processes is not important at high E/N values.

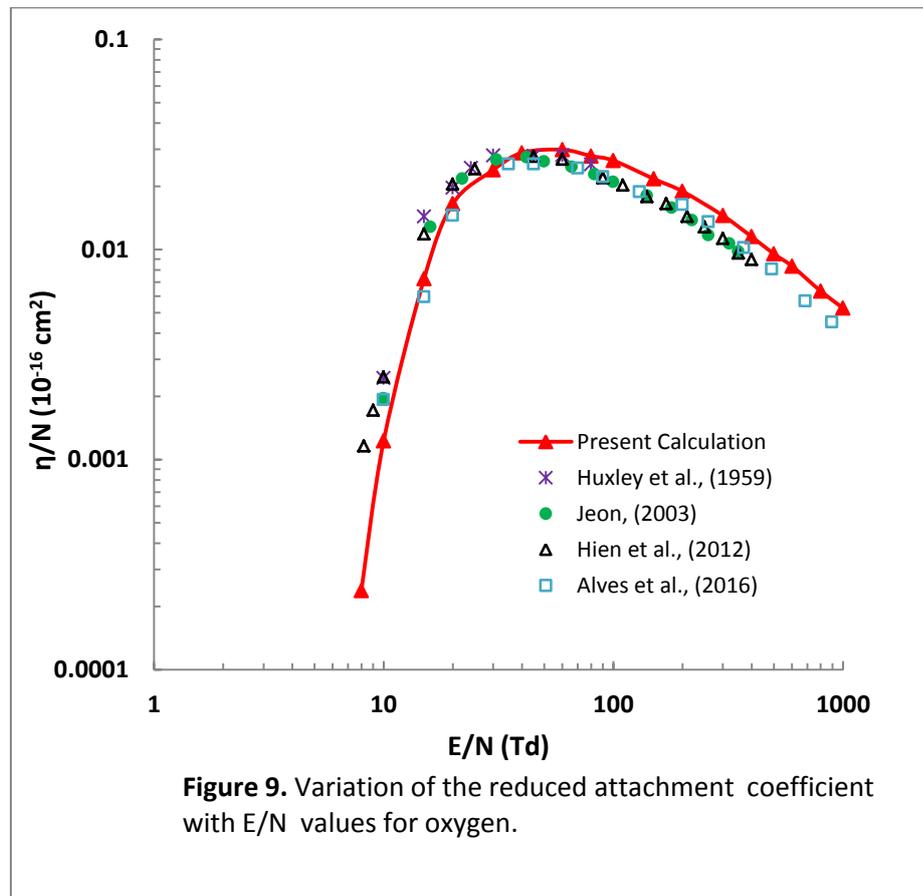
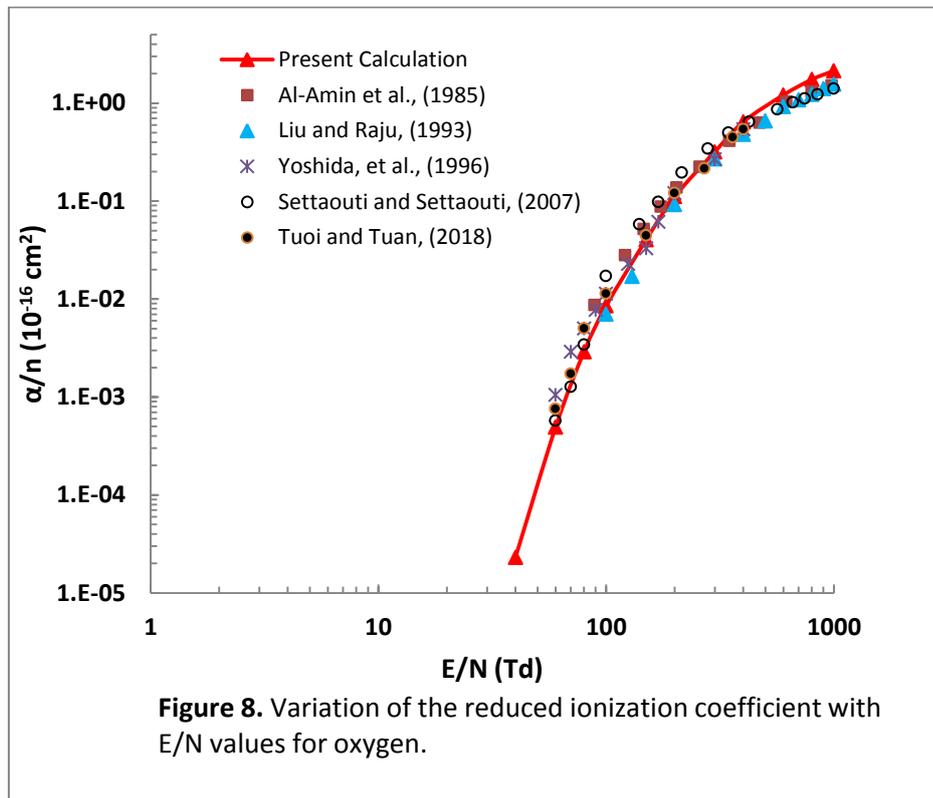
Figure (11) illustrates the percentage energy losses by elastic and inelastic processes. At high $E/N=600$ Td, 25% energy lost through ionizing collisions, 74% to electronic excitation, and 1% to attachment collisions. While at Lowest E/N ($E/N=1$ Td), 10% energy lost through momentum transfer (elastic) collisions and 90% to vibrational collisions. Maxima for vibrational excitation loss occurs at $E/N= 2$ Td. At high E/N the energy transfer to attachment is very small, only electronic and ionization fraction are dominants.

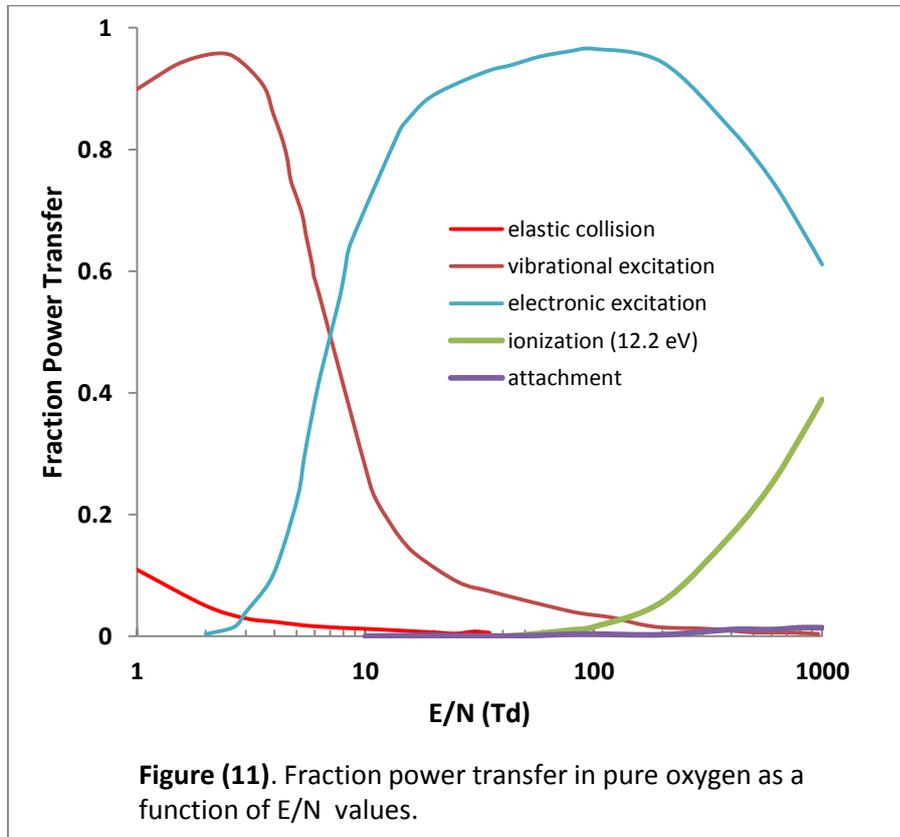
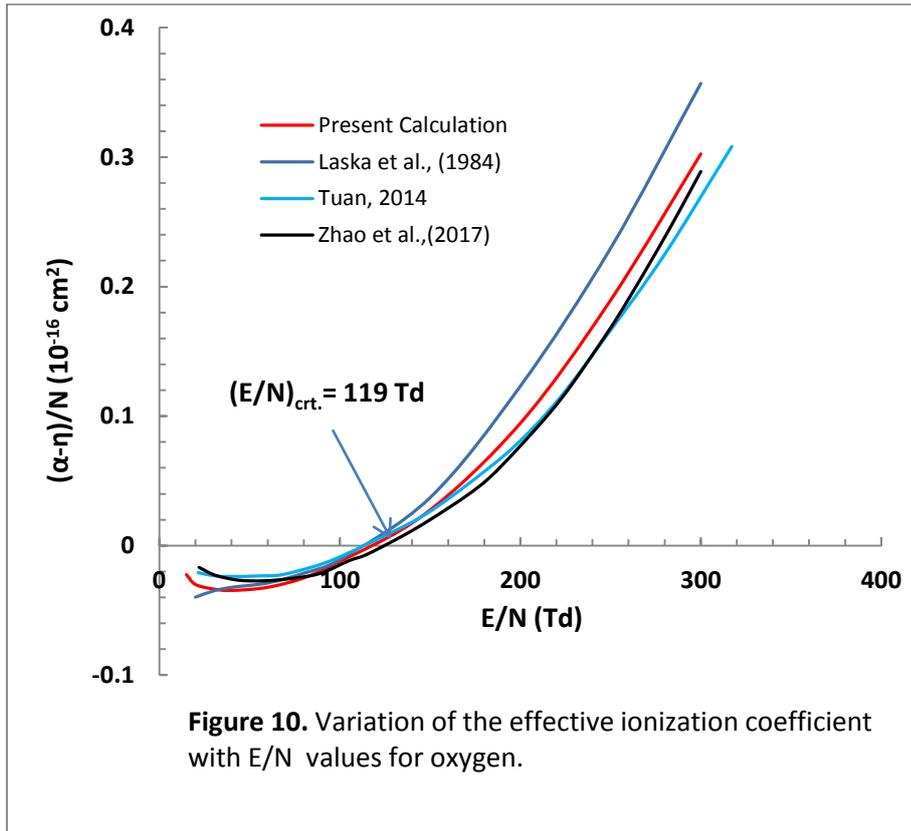












4. Conclusion

The electron swarm parameters in pure oxygen has been calculated and analyzed over a wide range of E/N varying from 0.1 Td to 1000 Td, using two-term solution of Boltzmann equation, where the effect of the ionization coefficient would be considered. A set of electron/molecule cross-sections has been used to calculate electron energy distribution function (EEDF) and swarm parameters namely, electron drift velocity, mean electron energy, characteristic energy, electron mobility, diffusion coefficient, ionization and attachment coefficient. The distribution function strongly effected by changing the electric field strength E/N, whereas, the effect of second kind collision (super-elastic collision) is not important at high E/N. The calculated swarm parameters are agree well with previous experimental and theoretical values. Moreover, the reduced critical electric field strength $(E/N)_{\text{cr}}$ is calculated using effective, ionization curves. Furthermore, the energy losses by different types of elastic and inelastic collision have been explained.

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