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دراسة نظرية لدالة توزيع طاقة الالكترون ومعامل التاين في غازي الكربتون والكلور النقيين وخليطيهما

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الملخص:

تم اجراء دراسة نظرية لدالة التوزيع الطاقي للالكترون ومعامل التاين الالكتروني لغازي Cl_2 و النقيين وخليطيهما من خلال معادلة الانتقال الطاقي لبولتزمان حسابيا من خلال البرنامج Kr (code-Kinema-Elendif), وقد تم مقارنة النتائج التي تم الحصول عليها مع نتائج باحثين اخرين و وجد توافقا جيدا بينهم. كما وتم استخدام المقاطع العرضية لكل من هاذين الغازين ووجد بأن دوال التوزيع هي لاماكسويلية و تمتلك اختلافات في الطاقة تعكس دور عمليات تبادل طاقة النقي يوثر على Kr الى غازCl_2الأكترون الجزيئي .وقد لوحظ ان اضافة نسبة قليلة من غاز دالة التوزيع الطاقي ومعامل التاين تاثيرا واضحا.

كلمات دالة: البلازما, دالة التوزيع الطاقي للألكترون, معادلة الأنتقال لبولتزمان,معامل التاين , تفريغ الغاز, الليزر الغازي.

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Theoretical study of the Electron energy distribution Function and the ionization coefficient for pure Krypton, Chlorine discharge and their mixtures

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ABSTRACT

Theoretical calculations performed on the electron energy distribution function (EEDF) and ionization coefficient (α/N) in pure Krypton (Kr) and pure chlorine (Cl_2) and their mixtures by using Boltzmann transport equation. The numerical solutions are utilized within the international computer code kinema-Elendif. The results have been compared successfully with other researcher's results and there was a good agreements. A set of electron cross-section data have been used in the calculation. The calculated distribution function found to be remarked non-Maxwellian that have energy variations which reflect the import electron-molecule energy exchange processes. It is noted that a

small quantity of Cl_2 add to pure Kr affects the electron energy distribution function and the ionization coefficient of the mixtures.

Keyword: *Plasma, Electron Energy Distribution Function (EEDF), Boltzmann transport equation, ionization coefficient, gas Discharges. Gas laser.*

1. Introduction

In order to gain more insights into the electron energy distribution function (EEDF) which is describe the motion of particles of plasma or gas considered as pure or as a mixture, one can say the electron energy distribution function(EEDF) is essential in plasma modeling because it is needed to compute reaction rates for electron collision reactions. Because electron transport properties can also be derived from the EEDF. Various approaches can be used to describe the EEDF, such as Maxwellian, Druyvesteyn, or using a solution of the Boltzmann equation [1].

The physics of electron swarms parameters (such as ionization coefficient which is a function of ionization probability, distribution function and drift velocity.) has been attracting considerable attention during the past decades because of the interest in the various physical effects taking place in these settings and by the need for accurate input data in discharge modeling. The numerical techniques and computational resources made it possible to gain more detail on physics of particle swarms. Most of the

intentions have been devoted to electron swarms, the electron energy distribution function and the transport properties have been determined for a wide variety ranges of gases and gas mixtures [2].

Using mixtures as a reactive medium with rare gases such as krypton has been a subject of great interest in DC electrical plasma discharge medium, because of their application in film etching and deposition in microelectronic device fabrications. Several industrial applications can be made using low-temperature plasmas such as the production of integrated circuits that involves deposition and etching [3, 4 and 5].

Krypton is used in tungsten-filament projection lamps, for home movies and slide projectors. A krypton gas laser produces a very intense and concentrated light. These lasers (531+ nm) are used for medical applications such as surgery on the retina of the eye [6].

The choice of Krypton as a host medium gas for interactions with the atoms and molecules of guest gases (as chlorine) depend on the availability of their energy cross-sections for elastic and inelastic type's interactions [7].

Gas mixtures containing molecular chlorine (Cl_2) are widely used as plasma processing gases for etching of semiconductors. The Cl_2 gas is also an important component of gas mixtures used for rare-gas halide lasers [8]. Moreover, Chlorine (Cl_2) has been selected as an available substitution candidate for the SF_6 gas in the published patent of Luly and

Richard [9] because of low global Warming potential (GWP) and high dielectric strength. This gas and its mixtures have been widely used in plasma etching of semiconductors, metals, and gate stacks with high-κ dielectrics and low-κ dielectric films [10]. Appending of that, chlorine is used in water disinfection and bleaching textiles. Chlorine also is used to manufacture other compounds ranging from pharmaceuticals to insecticides [4, 5 and 11]. For both atoms of chlorine molecular, electron collisions of the electric discharge with molecular chlorine were taken into account. The purpose is to investigate the effect of gas composition on the electron transport coefficients.

In this paper the EEDF and the ionization coefficient as a function of E/N range (ratio of the applied electric field *E* to the neutral number density *N*) in pure Kr, Cl_2 and Kr-Cl_2 mixtures were analyzed and calculated by using the numerical code Kinema-Elendif which has been written in FORTRAN 90/95, which solves the time-dependent Boltzmann equation for a given mixture of partially ionized gases with transport coefficients for pure gases and their mixture. This program has been developed by W. L Morgan and Kinema Research and software. It comprises over one hundred and eighty chemical reactions and uses as input parameters the initial concentrations of the primary gases calculates the electron energy distribution function by solving Boltzmann's equation in the time-dependent form in the plasma [12].

A comparison is made between a pure Kr and one with small quantities of (Cl_2 5% and Kr 20%) mixture and with (Cl_2 20% and Kr 80%).It noted that a small quantity of Cl_2 add to pure Kr is affects the EEDF of the mixture. In general, when the percentage ratio of the Cl_2 gas increases in the mixtures, the value of the EEDF and electron transport coefficient increase progressively to those of the pure Kr gas.

The calculated values are obtained by suitably averaging over the electron energy distribution function (EEDF), which is itself obtained by using the cross sections as input to a

Boltzmann solver.

Theory

A) Boltzmann equation

The general form of the Boltzmann equation is [3, 4 and 12]:

$$\left(\frac{\partial}{\partial t}+V.\nabla_r-\frac{eE}{m}.\nabla_V\right)f(r,v,t)=\left(\frac{\delta}{\delta t}\right)_{coll.} \quad \text{.....(1)}$$

Where (f) is the electron velocity distribution function (EEDF), (V) the electron velocity, (∇_r) is the special gradient in r-dimension, while(e/m) is the ratio of electron charge to its mass which is refers to the acceleration due to applied electric field (E) ($V.cm^{-1}$) and (∇_V) is the velocity-gradient operator. The term on the right hand side of equation (1)

is the collision integral, which accounts for electron energy transferred in elastic and inelastic collisions.

Solution of the transport Boltzmann equation for the electron – velocity distribution function $f\ (r\ ,v,t)$ is approximated by $f\ (\ v)$, since it is assumed that electron fields is independent of space and time and the problems of electron interactions are spatially uniform, so that the velocity dependence distribution function can be written by Legendre series expansion as follow:-

$$f(r,v,t) = f + \sum_{l=0}^{\infty} f_l(r,v,t)\, P_l(cos\theta) \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (2)$$

Or, in terms of the velocity dependence approximation

$$f(v) \approx f(v) + \sum_{l=0}^{\infty} f_l(v)\, P_l(cos\theta) \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (3)$$

The relation between the electron energy distribution function $f\ (\varepsilon)$ and mean energy in eV is:

$$\langle\varepsilon\rangle = \int_0^{\infty} \varepsilon^{3/2}\, f(\varepsilon) d\varepsilon \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots (4)$$

It is well known that the swarm parameters of electrons and collision cross-sections with molecules are related to each other through the medium of the velocity distribution function of the swarm. Therefore, the ionization coefficient can be calculated from [12]:

$$\frac{\alpha}{N} = \frac{1}{V_d}\left(\frac{2e}{m}\right)^{1/2}\int_0^{\infty}\sigma_i(\varepsilon)\, f(\varepsilon)d\varepsilon \ldots\ldots\ldots\ldots\ldots\ldots (5)$$

Where $(\frac{\alpha}{N})$ is the ionization coefficient of electron and (V_d) is the drift velocity, (m) is the electron mass and $\sigma_i(\varepsilon)$ is the ionization cross-section.

A) Cross sections

1-Krypton (Kr)

The set of excitation cross-section ($2P_5$ 3P (P_2)) for the metastable state, ionization cross-section in Krypton was taken from measured values of Sakai *et.al* [13]

2-Chlorine (Cl_2)

For chlorine gas, electron collisions with both atomic and molecular where taken into account. The purpose was to investigate the effect of gas composition on the electron transport because various kind of elastic processes such as excitation and ionization play an important role [5, 7 and 8].

The chlorine molecules in the metastable state $C^1\pi$ is dissociative excitation with a threshold of (3.12eV) and electronic excitations (molecular) to ($B^1\pi$, 2.49eV loss) and ($2^1\pi$ and $2^1\Sigma$, 9.25eV loss) and

electronic excitations (atomic) with threshold of (8.9, 10.4, 10.9, 11.8, 12.0 and 12.4) eV.

The vibrational excitation plays an important role at intermediate values of E/N with energy loss (0.0689) eV. Molecular ionization with energy loss of (11.47) eV and also atomic ionization with energy loss of (12.99) eV are given by [14].

2. Result and Discussion

The present study has resulted in a set of cross-sections for Kr and Cl_2 by using Boltzmann transport equation. The EEDF for pure Kr and pure Cl_2 and their mixture (Kr-Cl_2) for different concentrations [(95-5) % (Kr-Cl_2), (80-20) % (Kr-Cl_2)] at E/N=27Td, E/N=120Td have been plotted as a function of electron energy values and they are completely presented in figures (1 and 2), respectively.

In figures (1) and (2), the EEDF is affected strongly by changing the value of E/N and it can be note that EEDF for pure Krypton had longer tail (15eV) but a small quantity of chlorine (5%) modifies this function to (14eV) and also adding (20%) of chlorine modifies the function to (11.25eV), These processes led to a displacement for the tail of EEDF.

In a matter of fact, the accuracy of the calculation depends on the accuracy of the electron-molecule cross-section sets. Calculations of the

electron energy distribution function for discharge processes of Kr, Cl_2 and their mixtures have shown that the distribution is highly non-Maxwellian because The EEDF decreased when E/N increase, it is attributed to the electron collisions number that will increase with neutral molecules or atoms, this cause the electron energy losses. Also, some of electrons have very weak energy and this increases the probability of attachment with neutrals atoms to create negative ions.

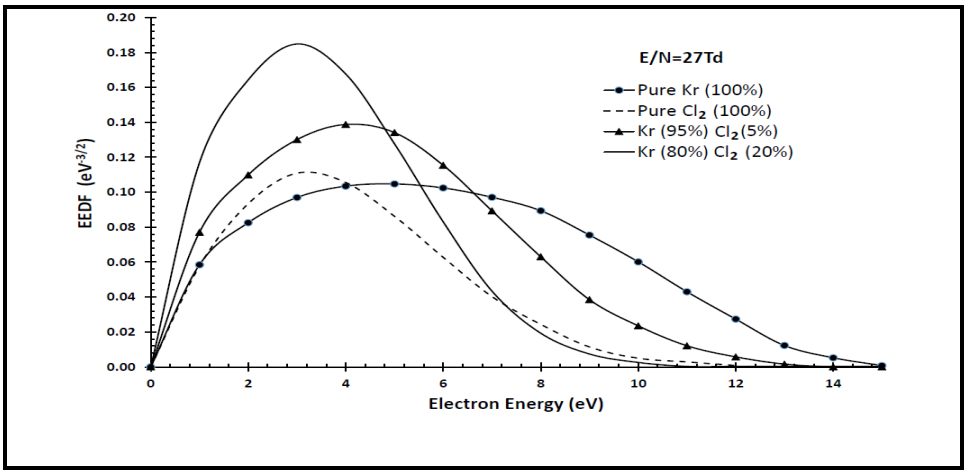

Figure 1 The electron energy distribution function as a function of electron energy in pure Kr, pure Cl_2 and (Kr-Cl_2) mixtures at E/N=27Td.

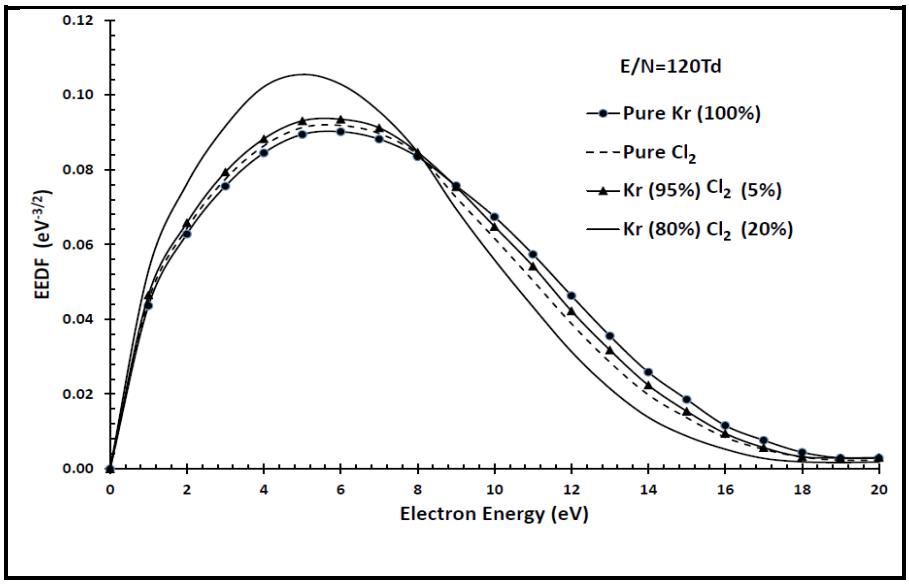

Figure 2 The electron energy distribution function as a function of electron energy in pure Kr, pure Cl_2 and (Kr-Cl_2) mixtures at E/N=120Td.

Table (1): The data of electron energy distribution function (EEDF) as a function of electron energy for pure Kr, pure Cl_2 and their mixtures at E/N=27Td.

Electron energy (eV)	EEDF ($eV^{-3/2}$)			
	Pure Kr	Pure Cl_2	Kr (95%)-Cl_2 (5%)	Kr (80%)-Cl_2(20%)
0.00	0.00	0.000	0.00	0.00
1.00	0.06	0.058	0.12	0.08
2.00	0.08	0.093	0.16	0.11
3.00	0.10	0.111	0.18	0.13
4.00	0.10	0.106	0.17	0.14

5.00	0.10	0.086	0.13	0.13
6.00	0.10	0.063	0.08	0.12
7.00	0.10	0.040	0.04	0.09
8.00	0.09	0.024	0.02	0.06
9.00	0.08	0.011	0.01	0.04
10.00	0.06	0.005	0.00	0.02
11.00	0.04	0.003	0.00	0.01
12.00	0.03	0.001	0.00	0.01
13.00	0.01	0.00	0.00	0.00
14.00	0.01	0.00	0.00	0.00
15.00	0.00	0.00	0.00	0.00

Table (2): The data of electron energy distribution function (EEDF) as a function of electron energy for pure Kr, pure Cl_2 and their mixtures at E/N=120Td.

Electron energy (eV)	EEDF ($eV^{-3/2}$)			
	Pure Kr	Pure Cl_2	Kr (95%)-Cl_2 (5%)	Kr (80%)-Cl_2(20%)
0.00	0.000	0.000	0.000	0.000
1.00	0.044	0.046	0.053	0.045
2.00	0.063	0.066	0.076	0.064
3.00	0.076	0.079	0.092	0.077

4.00	0.084	0.088	0.102	0.086
5.00	0.089	0.093	0.105	0.091
6.00	0.090	0.093	0.103	0.092
7.00	0.088	0.091	0.096	0.090
8.00	0.083	0.085	0.085	0.084
9.00	0.076	0.075	0.070	0.073
10.00	0.067	0.065	0.056	0.062
11.00	0.057	0.054	0.043	0.050
12.00	0.046	0.042	0.031	0.039
13.00	0.036	0.032	0.021	0.029
14.00	0.026	0.022	0.014	0.020
15.00	0.019	0.015	0.009	0.014
16.00	0.012	0.009	0.005	0.008
17.00	0.008	0.006	0.003	0.005
18.00	0.004	0.003	0.002	0.003
19.00	0.003	0.003	0.002	0.002
20.00	0.003	0.003	0.002	0.002

The ionization coefficient for each pure gas (Kr, Cl_2) and their mixtures as a function of E/N value are presented in figures (3 and 4), respectively.

The nonlinear behavior of ionization coefficient (□/N) which is calculated by eq. (5) is shown in figure 3. In this figure, the ionization coefficient for each pure gas is increase with increasing E/N value because the electrons acquire enough energy from the applied field to reach the ionization level, In this case, the number of energetic electrons

(which cause the ionization process) increase with increasing E/N values according to the ionization cross-section of each gases (Kr, Cl_2).The results of pure Kr are in a good agreement with experimental work measured by Moores (2001) and Wetzel (1987) also the results of pure Cl_2 are in a sensitive agreement with the experimental results measured by Sirvastava (1998) and theoretical results measured by Stevie (1982).

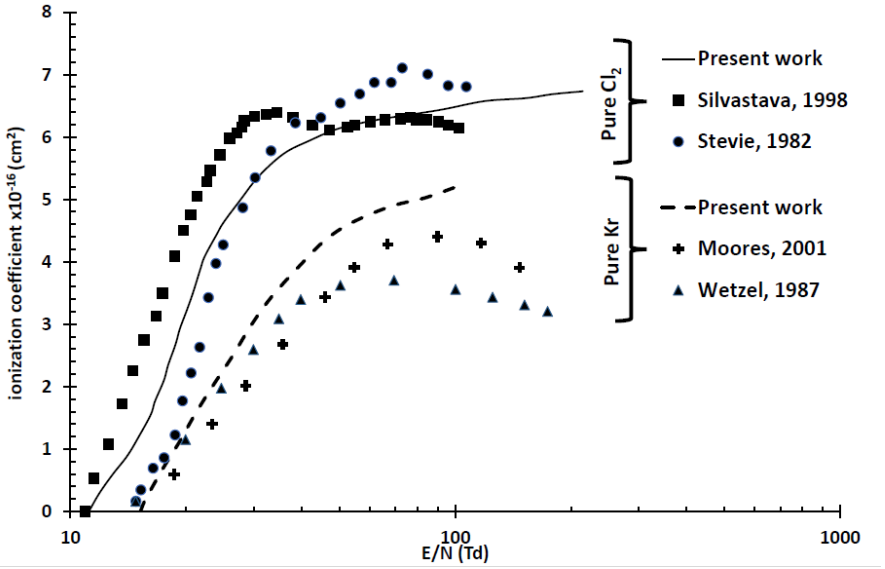

Figure 3 The ionization coefficient of electron as a function of E/N for pure (Kr, Cl_2).

The process of mixing (by inserting Chlorine gas to Krypton gas), causes a noticeable increase in the values of electron ionization coefficient accompanied by increasing in E/N value.

In figure (4), the ionization coefficient is increase with increasing Cl_2 concentration in the mixtures. It can be note that the ionization

coefficient with ratio (80-20) % (Kr-Cl_2) mixtures had a highest value($11.6x10^{-16}$ cm^2) than the ratio (95-5) % (Kr-Cl_2) mixtures which had value($6.8x10^{-16}$ cm^2) compared with the value of the pure krypton with value of ($5.2x10^{-16}$ cm^2). This behavior is due to the increase in the number of electron that causing the ionization also related to the stability of ionization state and the electronic excitation of Krypton. In other hand, the effect of elastic scattering take over the other processes due to the fact that pure Krypton gas had low energy transfer cross-section.

Table (3): The data of ionization coefficient as a function of E/N value for pure (Kr, Cl_2) of Moores(2001),Wetzel(1987),Sirvestava(1998),Stevie(1982) and present work.

Pure Kr	Pure Cl_2

E/N (Td)	Present work	E/N (Td)	Moores, 2001	E/N (Td)	Wetzel, 1987	E/N (Td)	Present work	E/N (Td)	Srivestava, 1998	E/N (Td)	Stevie, 1982
12.00	0.00	14.0	0.0	14.8	0.2	12.00	0.36	10.9	0.0	14.8	0.2
18.00	0.84	18.6	0.6	19.9	1.2	18.00	2.36	11.5	0.5	15.2	0.3
24.00	2.09	23.4	1.4	24.7	2.0	24.00	4.48	12.5	1.1	16.4	0.7
30.00	3.08	28.5	2.0	29.9	2.6	30.00	5.30	13.6	1.7	17.5	0.9
36.00	3.67	35.6	2.7	34.8	3.1	36.00	5.75	14.5	2.3	18.7	1.2
42.00	4.11	45.9	3.4	39.7	3.4	42.00	5.95	15.5	2.7	19.6	1.8
48.00	4.45	54.7	3.9	50.3	3.6	48.00	6.11	16.7	3.1	20.6	2.2
54.00	4.64	66.8	4.3	69.4	3.7	54.00	6.20	17.3	3.5	21.7	2.6
60.00	4.78	90.0	4.4	100.2	3.6	60.00	6.27	18.7	4.1	22.8	3.4
66.00	4.86	116.8	4.3	125.1	3.4	66.00	6.31	19.6	4.5	23.9	4.0
72.00	4.94	146.9	3.9	151.5	3.3	72.00	6.34	20.5	4.8	24.9	4.3
78.00	4.98	-----	-----	173.9	3.2	78.00	6.38	21.3	5.1	28.0	4.9
84.00	5.05	-----	-----	14.8	0.2	84.00	6.42	22.6	5.3	30.2	5.3
90.00	5.11	-----	-----	-----	-----	90.00	6.44	23.1	5.5	33.2	5.8
96.00	5.17	-----	-----	-----	-----	96.00	6.48	24.5	5.7	38.4	6.2
102.00	5.23	-----	-----	-----	-----	102.00	6.52	26.0	6.0	44.7	6.3
-----	-----	-----	-----	-----	-----	108.00	6.55	27.1	6.1	50.3	6.5
-----	-----	-----	-----	-----	-----	114.00	6.58	27.9	6.2	56.5	6.7
-----	-----	-----	-----	-----	-----	120.00	6.59	28.3	6.3	61.6	6.9
-----	-----	-----	-----	-----	-----	126.00	6.61	30.0	6.3	68.2	6.9
-----	-----	-----	-----	-----	-----	132.00	6.61	32.3	6.4	72.8	7.1
-----	-----	-----	-----	-----	-----	138.00	6.63	34.5	6.4	84.8	7.0
-----	-----	-----	-----	-----	-----	144.00	6.63	37.9	6.3	95.9	6.8
-----	-----	-----	-----	-----	-----	150.00	6.64	42.5	6.2	106.9	6.8

-----	-----	-----	-----	-----	-----	156.00	6.64	47.0	6.1	-----	-----
-----	-----	-----	-----	-----	-----	162.00	6.66	52.5	6.2	-----	-----
-----	-----	-----	-----	-----	-----	168.00	6.67	54.8	6.2	-----	-----
-----	-----	-----	-----	-----	-----	174.00	6.69	60.2	6.2	-----	-----
-----	-----	-----	-----	-----	-----	180.00	6.70	65.7	6.3	-----	-----
-----	-----	-----	-----	-----	-----	186.00	6.72	72.2	6.3	-----	-----
-----	-----	-----	-----	-----	-----	192.00	6.72	76.5	6.3	-----	-----
-----	-----	-----	-----	-----	-----	12.00	0.36	79.3	6.3	-----	-----
-----	-----	-----	-----	-----	-----	-----	-----	84.1	6.3	-----	-----
-----	-----	-----	-----	-----	-----	-----	-----	90.4	6.2	-----	-----

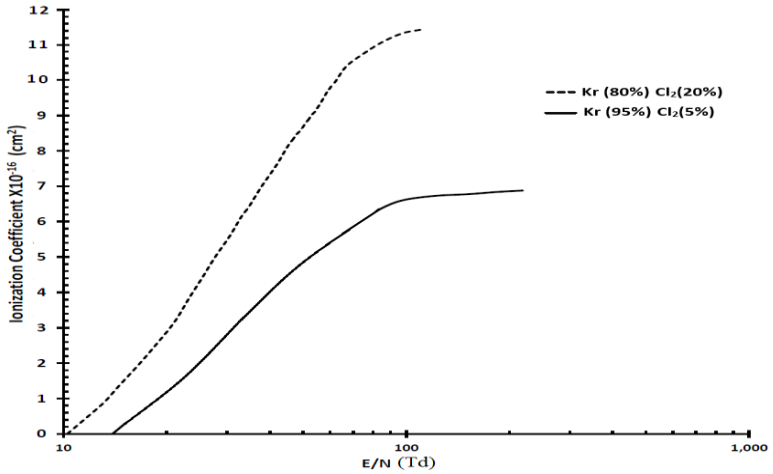

Figure 4 The ionization coefficient of electron as a function of E/N for (Kr-Cl_2) (80:20) %, (95:5) % gas mixtures.

Table (4): The data of the ionization coefficient of electron as a function of E/N for (Kr-Cl_2) (80:20) %, (95:5) % gas mixtures.

E/N (Td)	Kr (80%)-Cl_2(20%)	Kr (95%)-Cl_2(5%)	E/N (Td)	Kr(80%)-Cl_2(20%)	Kr(95%)-Cl_2(5%)
10.00	-----	0.00	112.00	6.66	11.45
16.00	0.27	1.75	118.00	6.66	-----
22.00	1.51	3.41	124.00	6.69	-----
28.00	2.57	5.09	130.00	6.71	-----
34.00	3.41	6.38	136.00	6.71	-----
40.00	3.99	7.37	142.00	6.73	-----
46.00	4.56	8.26	148.00	6.75	-----
52.00	5.00	8.97	154.00	6.75	-----
58.00	5.36	9.65	160.00	6.77	-----
64.00	5.62	10.21	166.00	6.77	-----
70.00	5.89	10.56	172.00	6.80	-----
76.00	6.11	10.83	178.00	6.82	-----
82.00	6.33	11.00	184.00	6.82	-----
88.00	6.46	11.16	190.00	6.84	-----
94.00	6.55	11.27	196.00	6.84	-----
100.00	6.62	11.38	200.00	6.84	-----
106.00	6.64	11.40	-----	-----	-----

3. Conclusion

By using the numerical Kinema-Elendif code, it was possible to investigate the behavior of the EEDF for pure Krypton and pure Chlorine and with their mixtures. The results have been compared with experimental and theoretical work of other researchers and there was a good agreements between them. The code was used to compare a pure Krypton, a [(95-5) % (Kr-Cl_2), (80-20) % (Kr-Cl_2)] mixture.

In general, it can say that for a given electric field, □/N increases with decreasing rare gas percentage in the mixtures and with the rare gas ionization potential. The electron energy distribution function of the mixture with the rare gas at higher ionization potential will be more populated at higher electron energies. Consequently, more introduce of Cl_2 ionization contribution to the total □/N will increase the rare gas ionization potential. The results have shown that the calculated□□/N, where in good agreement with computational works of other researchers.

Finally, when the percentage ratio of the Cl_2 gas in binary mixtures increases, the values of the electron energy distribution function and electron transport coefficients increase to those of the pure Cl_2. The main advantage of Kr-Cl_2 mixtures is to reach the ionization state with less electron energy. Therefore, the optimum case in this study is 20% Cl_2

which add to 80% Kr which indicates to the advantage of this concentration ratio in the used mixture.

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