Analysis of Electron Transport Coefficients in CF_3I-N_2 Mixture Gas Using an Electron Swarm Study

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Abstract. The consistent electron transport coefficients, for not only pure atoms and molecules but also binary gas mixtures, are necessary to quantitatively understand plasma phenomena and ionized gases. Electron transport coefficients in CF_3I-N_2 mixture gas, therefore, were calculated and analyzed using a two-term approximation of the Boltzmann equation in the E/N range (ratio of the electric field E to the neutral number density N) of 10 - 1000 Td (1 Td = 10^{-17} V.cm^2) for the first time. These coefficients include electron drift velocity, density-normalized longitudinal diffusion and density-normalized effective ionization coefficients. The present results were in good agreement with the available experimental data over a wide range of E/N. The limiting field strength value of E/N for 70% CF_3I-N_2 mixture was derived and greater than that of the pure SF_6 gas. Gas mixtures of 65 – 75% CF_3I-N_2 could be considered to use in high voltage and many industries.

Keywords: Trifluoroiodomethane, Boltzmann equation analysis, electron transport coefficients, mixture gas.

1 Introduction

Sulfur hexafluoride (SF_6) has been widely used as an isolated gas in high voltage equipment. The Kyoto Protocol, however, has listed the greenhouse gases as CO_2, CH_4, N_2O, hydrofluorocarbons (HFCs), perfluorocarbons (PFCs) and SF_6, and we need to regulate the emissions and utilizations of those gases in the many industries [1]. Recently, much research has been concentrated on trifluoroidomethane (CF_3I) gas because of its low global warming potential, very short atmospheric lifetime and relatively low toxicity gas [2]-[4]. It is a gas that is a substitution candidate for the SF_6 gas and as a candidate to the replacement of potent greenhouse affects. This gas has also been considered to be a candidate replacement for bromotrifluoromethane (CF_3Br), which is used in aircraft for fuel inerting and for fire-fighting [2]. The boiling point of CF_3I gas is higher than that of the SF_6 gas. At an absolute pressure of 0.5 MPa, CF_3I becomes liquids at about 26 ^{0}C, whereas the SF_6 gas becomes liquids at about -30 ^{0}C. On the other hand, the SF_6 gas is used in gas circuit breakers at 0.5 to

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0.6 MPa. Therefore, it is impossible to use CF_3I gas, if this gas is used at this pressure level [3]. However, in order to reduce the liquefaction temperature of CF_3I gas, Taki *et al.* [3] decreased partial pressure by mixing it with other gases such as N_2 and CO_2. For example, the boiling point can be reduced from about 26 ^{0}C (pure CF_3I) to about -12 ^{0}C at 0.5 MPa by using a 30% CF_3I-CO_2 mixture [4]. Therefore, it is necessary to mix the CF_3I gas with different buffer gases.

Recently, the electron transport coefficients in CF_3I-N_2 [5], [6] mixtures were measured and calculated for the entire concentration range of CF_3I percentages and in the E/N ranges (ratio of the electric field E to the neutral number density N) of about 100 - 600 Td (1 Td = 10^{-17} V.cm^2). In general, the electron transport coefficients for molecules and binary mixture gases are necessary for quantitative understanding of plasma phenomena. Moreover, the collision processes and electron transport coefficients of the binary mixtures of CF_3I gas with other gases such as Xe, Ne, He, N_2, CO_2, and O_2 have been scarce so far.

In the present study, in order to gain more insight into the electron transport coefficients, the electron drift velocity, density-normalized longitudinal diffusion coefficient, and density-normalized effective ionization coefficient in the E/N range of 10 - 1000 Td and the limiting field strength of E/N, $(E/N)_{lim}$, for CF_3I-N_2 mixtures were calculated using a two-term approximation of the Boltzmann equation for the energy. The calculated electron transport coefficients in the CF_3I-N_2 mixtures with CF_3I concentrations between 5% and 70% were compared with the recent experimental results [5]. The negative differential conductivity (NDC) phenomena, that is, decreasing electron drift velocity with increasing electric field strength, in these binary gas mixtures were suggested. The present electron transport coefficients calculated were also compared with those of pure SF_6 gas in the experiments. The binary mixtures CF_3I-N_2 with CF_3I concentration equal to about 65 - 75% are considered to use in high voltage and many industries.

2 Analysis

In the present study, to the best of our knowledge, CF_3I molecule has the inelastic cross sections, which are comparable to and less than the elastic momentum transfer cross section at low energies and also at high energies [7]. We, therefore, chose a two-term approximation of the Boltzmann equation for calculating the electron transport coefficients in CF_3I-N_2 mixture gas. The electron transport coefficients were calculated using a two-term approximation of the Boltzmann equation for the energy given by Tagashira *et al.* [8] and gaseous electron collision cross section sets. The present two-term approximation of the Boltzmann equation was previously used for the Cl_2 [9] and TEOS [10] molecules. The briefly description for calculating the electron transport coefficients was also following represented. The electron drift velocity calculated from the solution of electron energy distribution function, f(ε, E/N), of the Boltzmann equation is defined as [11]

$$W = -\frac{1}{3}\left(\frac{2}{m}\right)^{1/2}\frac{eE}{N}\int_0^\infty \frac{\varepsilon}{q_m(\varepsilon)}\frac{df(\varepsilon, E/N)}{d\varepsilon}d\varepsilon. \quad (1)$$

where ε is the electron energy, m is the electron mass, e is the elementary charge, and $q_m(\varepsilon)$ is the momentum-transfer cross section.

The density-normalized longitudinal diffusion coefficient is defined as [12]

$$ND_L = \frac{V_1}{3N}\left(E\int_0^\infty \frac{\varepsilon}{q_T}\frac{\partial}{\partial\varepsilon}(F_1\varepsilon^{-1/2})d\varepsilon + \int_0^\infty \frac{\varepsilon^{1/2}}{q_T}F_0 d\varepsilon \right) - \left(\varpi_0 A_2 - \varpi_1 A_1 - \varpi_{02}\right). \quad (2)$$

where V_1 is the speed of electron, q_T is the total cross section, here F_n and ϖ_n (n = 0, 1, 2) are respectively the electron energy distributions of various orders and their eigenvalues. V_1, ϖ_n, ϖ_{0n}, and A_n are given by

$$V_1 = \left(\frac{2e}{m}\right)^{1/2};\ \varpi_0 = V_1 N\int_0^\infty \varepsilon^{1/2} q_i F_0 d\varepsilon;\ \varpi_1 = -\frac{V_1 E}{3N}\int_0^\infty \frac{\varepsilon}{q_T}\frac{\partial}{\partial\varepsilon}(F_0\varepsilon^{-1/2})d\varepsilon + (\varpi_0 A_1 - \varpi_{01});$$

$$\varpi_{0n} = V_1 N\int_0^\infty \varepsilon^{1/2} q_i F_n d\varepsilon;\ A_n = \int_0^\infty F_n d\varepsilon.$$

where q_i is the ionization cross section.

The Townsend first ionization coefficient is defined as [13]

$$\alpha/N = \frac{1}{W}\left(\frac{2}{m}\right)^{1/2}\int_I^\infty f(\varepsilon, E/N)\varepsilon^{1/2} q_i(\varepsilon)d\varepsilon. \quad (3)$$

where I is the ionization onset energy and $q_i(\varepsilon)$ is the ionization cross section.

The electron attachment coefficient is defined as [13]

$$\eta/N = \frac{1}{W}\left(\frac{2}{m}\right)^{1/2}\int_0^\infty f(\varepsilon, E/N)\varepsilon^{1/2} q_a(\varepsilon)d\varepsilon. \quad (4)$$

where $q_a(\varepsilon)$ is the attachment cross section. The electron collision cross sections for CF_3I determined by Kimura and Nakamura [7], [14] and N_2 determined by Nakamura [14] were used throughout the present study. The accuracy of the electron collision cross section set for each gas was confirmed to be consistent with all electron transport coefficients in each pure gas. A brief summary of several reaction processes of these gases for plasma modeling is given by some published reports and is listed in Table 1.

3 Results and Discussions

3.1 Electron Drift Velocities

The results for the electron drift velocities, W, as functions of E/N for the CF_3I-N_2 mixture gas calculated in the E/N range 10 < E/N < 1000 Td using a two-term approximation of the Boltzmann equation are shown in Fig. 1. The good agreements between the present results and those measured by Urquijo *et al.* [5] in Fig. 1(b) are satisfactory over the entire E/N range. Slight regions of the NDC phenomena in these gas mixtures are observed in the E/N range 15 < E/N < 170 Td. The NDC is relatively

shallow for all mixtures. The occurrences of these phenomena are due to the Ramsauer-Townsend minimum (RTM) of the elastic momentum transfer cross sections of the CF_3I molecule. The values of W are suggested to be between those of the pure gases over E/N > 100 Td and these values grow linearly over E/N > 200 Td. For the sake of comparison, the electron drift velocity obtained by Aschwanden [15] for the pure SF_6 gas is shown in Fig. 1(a). The calculated electron drift velocity in 70% CF_3I-N_2 in the E/N range of E/N < 400 Td is very close to that of the pure SF_6 gas.

Table 1. Reaction processes with threshold energy for plasma modeling in gases

Types of Collision	Reactions	Threshold Energies (eV)	References
* in CF_3I molecule			
Momentum transfer	$CF_3I + e \rightarrow CF_3I + e$	0	[7]
Vibrational excitation (3)	$CF_3I + e \rightarrow CF_3I(v=1, 2, 3) + e$	0.032 (v=1), 0.067 (v=2), 0.134 (v=3)	[7]
Electronic excitation	$CF_3I + e \rightarrow CF_3 + I + e$	4.7	[7]
Electronic excitation	$CF_3I + e \rightarrow CF_3 + I^* + e$	7.2	[7]
Electronic excitation	$CF_3I + e \rightarrow CF_2I + F + e$	7.9	[7]
Electronic excitation (2)	$CF_3I + e \rightarrow CF_2 + IF + e$	8.8, 9.6	[7]
Ionization	$CF_3I + e \rightarrow CF_3I^+ + 2e$	10.2	[7]
Dissociative attachment			[7]
+ zero-energy process	$CF_3I + e \rightarrow I^- + CF_3$		
+ 3.8 eV process	$CF_3I + e \rightarrow CF_2I + F^-$		
	$CF_3I + e \rightarrow CF_3^- + I$		
* in N_2 molecule			
Momentum transfer	$N_2 + e \rightarrow N_2 + e$	0	[14]
Vibrational excitation (7)	$N_2 + e \rightarrow N_2(v=1, 2, 3, 4, 5, 6, 7) + e$	0.288 (v=1), 1.76 (v=2), 1.91 (v=3), 2.07 (v=4), 2.09 (v=5), 2.17 (v=6), 2.18 (v=7)	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^*(A^3\Sigma_u^+) + e$	6.169	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^*(B^3\Pi_g) + e$	7.353	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^*(B'^3\Sigma_u^-) + e$	8.165	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^*(a^1\Pi_g) + e$	8.549	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^* + e$	9.756	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^*(C^3\Pi_u) + e$	11.032	[14]
Electronic excitation	$N_2 + e \rightarrow N_2^* + e$	12.579	[14]
Ionization	$N_2 + e \rightarrow N_2^+ + 2e$	15.5	[14]

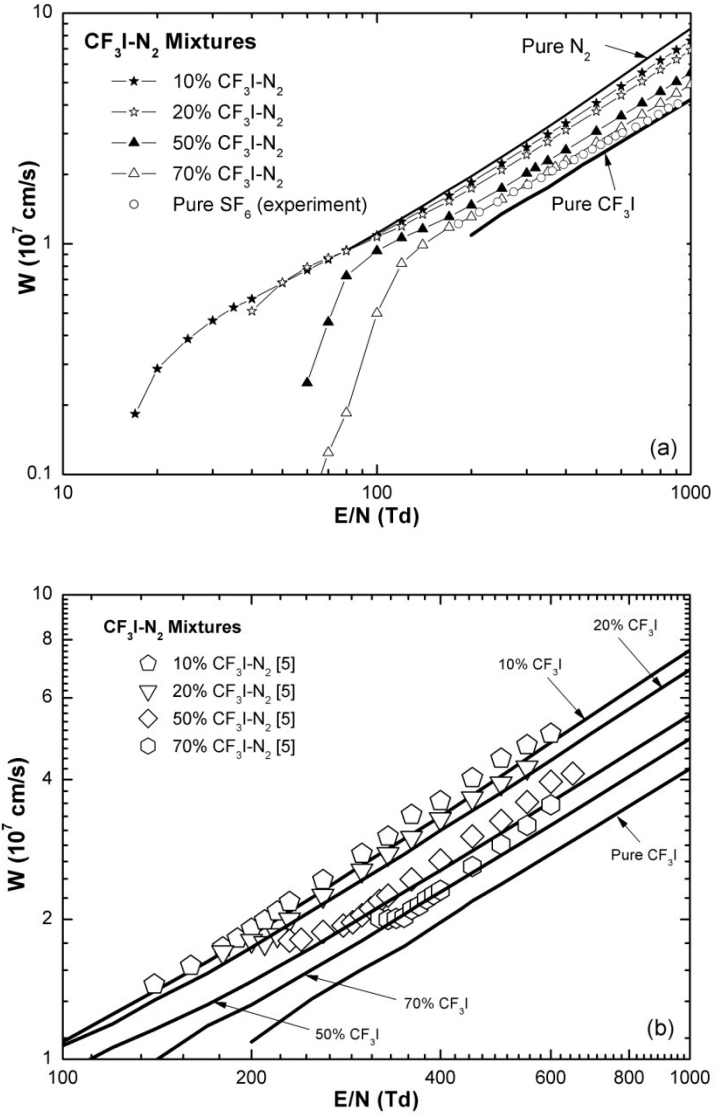

Fig. 1. Electron drift velocity, W, as functions of E/N for the CF_3I-N_2 mixtures with 10%, 20%, 50%, and 70% CF_3I. (a) The solid line and symbols show present W values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-N_2 mixtures. The solid curves show present W values calculated for the pure CF_3I and N_2 molecules. The open circle symbol shows the measurement value of the pure SF_6 [15]. (b) The figure shows comparisons between the results calculated and experiments [5] for the CF_3I-N_2 mixtures with 10%, 20%, 50%, and 70% CF_3I, respectively. The symbols show the experiment results of the CF_3I-N_2 mixtures [5]. The solid curves show the present W values calculated using a two-term approximation of the Boltzmann equation for the pure CF_3I and CF_3I-N_2 mixtures.

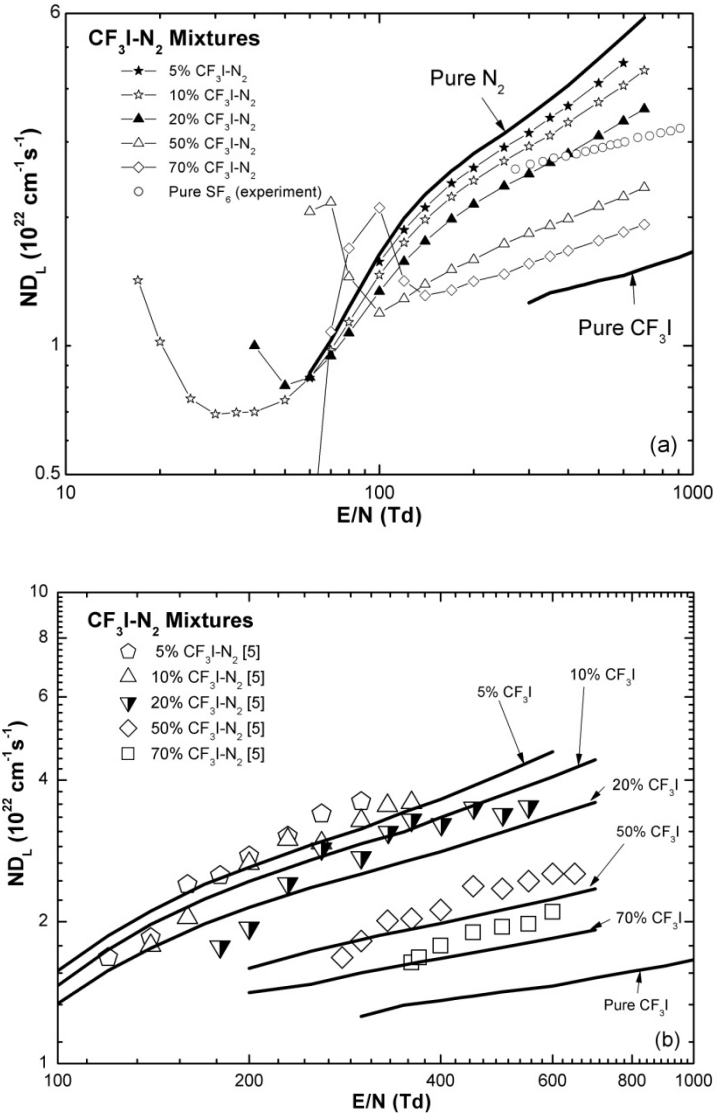

Fig. 2. Density-normalized longitudinal coefficient, ND_L, as functions of E/N for the CF_3I-N_2 mixtures with 5%, 10%, 20%, 50%, and 70% CF_3I. (a) The solid line and symbols show present ND_L values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-N_2 mixtures. The solid curves show present ND_L values calculated for the pure CF_3I and N_2 molecules. The open circle symbol shows the measurement value of the pure SF_6 [15]. (b) The figure shows comparisons between the results calculated and experiments [5] of the CF_3I-N_2 mixtures with 5%, 10%, 20%, 50%, and 70% CF_3I, respectively. The symbols show the experiment results of the CF_3I-N_2 mixtures [5]. The solid curves show the present W values calculated using a two-term approximation of the Boltzmann equation for the pure CF_3I, pure N_2, and CF_3I-N_2 mixtures.

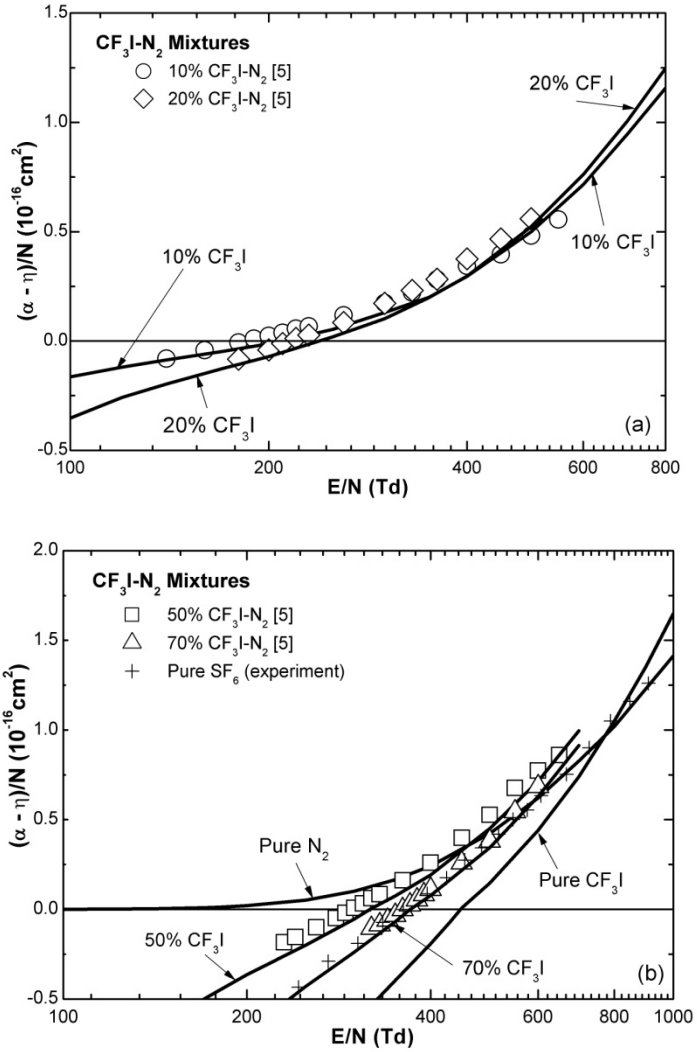

Fig. 3. (a), (b) Density normalized effective ionization coefficient, (α - η)/N, as functions of E/N for the CF_3I-N_2 mixtures with 10%, 20%, 50%, and 70% CF_3I. The solid curves show present (α - η)/N values calculated using a two-term approximation of the Boltzmann equation for the pure CF_3I, pure N_2 molecules, and CF_3I-N_2 mixtures. The cross symbol shows the measurement value of the pure SF_6 [15]. The symbols show the experiment results of the CF_3I-N_2 mixtures [5].

3.2 Density-Normalized Longitudinal Diffusion Coefficients

The results for the density-normalized longitudinal diffusion coefficients, ND_L, as functions of E/N for the CF_3I-N_2 mixture gas calculated in the E/N range 10 < E/N < 1000 Td using a two-term approximation of the Boltzmann equation are shown in

Fig. 2. In these binary mixtures, the values of ND_L are suggested to be between those of the pure gases over E/N > 200 Td. The agreements between the results calculated and those measured by Urquijo *et al.* [5] are satisfactory over the entire E/N range for the CF_3I-N_2 mixtures in Fig. 2(b). The density-normalized longitudinal diffusion coefficient for the pure SF_6 obtained by Aschwanden [15] is also shown in Fig. 2(a) for the aim of comparison. The ND_L values of the pure SF_6 are greater than those of these binary mixtures. The increased trends of values calculated for 50% and 70% CF_3I-N_2 mixtures are the same as those of the pure SF_6 gas with increasing electric field strength.

3.3 Density-Normalized Effective Ionization Coefficients

The results for the density-normalized effective ionization coefficients, $(\alpha - \eta)/N$, as functions of E/N for the CF_3I-N_2 mixture gas calculated using a two-term approximation of the Boltzmann equation are shown in Fig. 3. In these binary mixtures, the values of $(\alpha - \eta)/N$ are also suggested to be between those of the pure gases. The best agreements between the present results and those measured by Urquijo *et al.* [5] in Fig. 3(b) are satisfactory over the entire E/N range. For the sake of comparison, the density-normalized effective ionization coefficient obtained by Aschwanden [15] for the pure SF_6 gas is also shown in Fig. 3(b). The $(\alpha - \eta)/N$ values for 70% CF_3I-N_2 mixture are very close to those of the pure SF_6 gas over E/N < 550 Td.

Because of the accuracy of the electron collision cross sections for the present gases and the validity of the Boltzmann equation, the present results calculated are reliable over the E/N < 100 Td and the results have been also calculated for the first time. More experiments of the electron transport coefficients for the binary mixtures of the CF_3I gas with these buffer gases need to be performed over the wide range of E/N in the future. In general, when the percentage ratio of the CF_3I gas in binary mixtures increases, the values of the electron transport coefficients increase progressively to those of the pure CF_3I.

3.4 Limiting Field Strength Values of E/N

The limiting field strength values of E/N, $(E/N)_{lim}$, at which $\alpha = \eta$ for the CF_3I-N_2 mixture gas are derived at 133.322 Pa as shown in Fig. 4. The present $(E/N)_{lim}$ values calculated for the CF_3I-N_2 mixtures were in good agreement with those derived from measurement of Urquijo *et al.* [5] as shown in Fig. 4. These values are also compared with those of the SF_6-N_2 mixture gas [16] as shown in Fig. 4. The $(E/N)_{lim}$ values of the CF_3I-N_2 mixture are greater than that of the binary mixtures of CF_3I gas with the other gases over the entire CF_3I concentration. The $(E/N)_{lim}$ values of the SF_6-N_2 mixtures are the greatest when the SF_6 concentration increases to 50% in the binary mixtures. However, the $(E/N)_{lim}$ values of the CF_3I-N_2 mixtures are the greatest when the CF_3I concentration is greater than about 50% in the binary mixtures. It may be considered as a prospective substitute for the SF_6 gas. In Fig. 4, the 65 - 75% CF_3I-N_2 mixture gases are considered to use in high voltage and many industries if other chemical, physical, electrical, thermal, and economical studies are considered thoroughly.

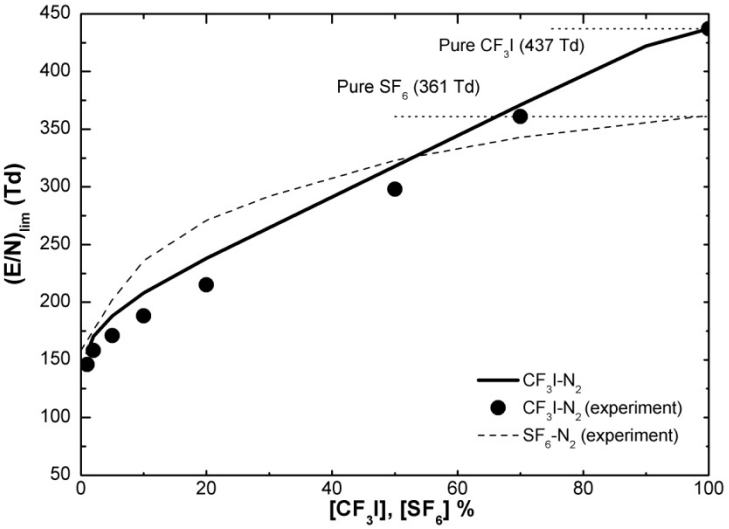

Fig. 4. Limiting field strength of E/N, $(E/N)_{lim}$, as a function of the percentage of CF_3I gas for the CF_3I-N_2 mixture. The solid line shows present $(E/N)_{lim}$ values for the CF_3I-N_2 mixture calculated using a two-term approximation of the Boltzmann equation. The solid symbols and dashed curve respectively show experimental $(E/N)_{lim}$ values for the CF_3I-N_2 [5] and SF_6-N_2 [16] mixtures.

4 Conclusion

The electron drift velocity, density-normalized longitudinal diffusion coefficient, and density-normalized effective ionization coefficient in the CF_3I-N_2 mixture gases are calculated using a two-term approximation of the Boltzmann equation for the energy in the E/N range of 10 - 1000 Td. The calculated electron transport coefficients in the CF_3I-N_2 mixtures are in good agreement with the experiments. The present results have been also calculated for the first time in the E/N range of 10 – 100 Td. The NDC phenomena in these binary gas mixtures are suggested. The electron transport coefficients calculated are also compared with those of the pure SF_6 gas in experiments. Moreover, the limiting field strength values of E/N for the 70% CF_3I-N_2 mixture gas are determined and essentially greater than those of the pure SF_6 gas. The binary mixtures of 65 - 75% CF_3I-N_2 are considered to use in high voltage and many industries. For the purposes of justification of the accuracy of our results, more experimental data for electron transport coefficients for the binary mixtures of CF_3I with these gases need to be performed over a wide range of E/N.

References

1. Conference of the Parties, Kyoto Protocol to the United Nations Framework Convention on Climate Change, Third Session Kyoto, Kyoto, Japan (1997)
2. Li, Y., Patten, O., Youn, D., Wuebbles, D.J.: Potential impacts of CF_3I on ozone as a replacement for CF_3Br in aircraft applications. Atmos. Chem. Phys. 6(12), 4559–4568 (2006)
3. Taki, M., Maekawa, D., Odaka, H., Mizoguchi, H., Yanabu, S.: Interruption capability of CF_3I gas as a Substitution candidate for SF_6 gas. IEEE Trans. Dielectrics and Elec. Insulation 14(2), 341–346 (2007)
4. Takagari, H., Kasuya, H., Mizoguchi, H., Yanabu, S.: Investigation of the performance of CF_3I gas as a possible substitute for SF_6. IEEE Trans. Dielectrics and Elec. Insulation 15(5), 1424–1429 (2008)
5. de Urquijo, J., Juárez, A.M., Basurto, E., Hernández-Ávila, J.L.: Electron impact ionization and attachment drift velocities and longitudinal diffusion in CF_3I and CF_3I-N_2 mixtures. J. Phys. D: Appl. Phys. 40(7), 2205–2209 (2007)
6. Deng, Y.K., Xiao, D.M.: The effective ionization coefficients and electron drift velocities in gas mixtures of CF_3I with N_2 and CO_2 obtained from Boltzmann equation analysis. Chin. Phys. B 22(3), 035101-1–6 (2013)
7. Kimura, M., Nakamura, Y.: Electron swarm parameters in CF_3I and a set of electron collision cross sections for the CF_3I molecule. J. Phys. D: Appl. Phys. 43(14), 145202-1–6 (2010)
8. Tagashira, H., Sakai, Y., Sakamoto, S.: The development of electron avalanches in argon at high E/N values. II. Boltzmann equation analysis. J. Phys. D: Appl. Phys. 10(7), 1051–1063 (1977)
9. Tuan, D.A., Jeon, B.H.: Electron collision cross sections for the Cl_2 molecule from electron transport coefficients. J. Phys. Soc. Jpn. 80(8), 084301-1–5 (2011)
10. Tuan, D.A., Jeon, B.-H.: Electron Collision Cross Sections for the Tetraethoxysilane Molecule and Electron Transport Coefficients in Tetraethoxysilane-O_2 and Tetraethoxysilane-Ar Mixtures. Journal of the Physical Society of Japan 81(6), 064301-1–8 (2012)
11. Huxley, L.G.H., Crompton, R.W.: The Diffusion and Drift of Electrons in Gases, ch. 6 and ch. 13. John Wiley & Sons, New York (1974)
12. Jeon, B.H.: Determination of electron collision cross-sections for the oxygen molecule by using an electron swarm study. J. Korean Phys. Soc. 43(4), 513–525 (2003)
13. Christophorou, L.G., Hunter, S.R.: From basic research to application. In: Christophorou, L.G. (ed.) Electron-Molecule Interations and Their Applications, vol. 2, pp. 318–412. Academic Press, Florida (1984)
14. Nakamura, Y.: private communication. Tokyo Denki Univ., Tokyo (November 2010)
15. Aschwanden, T.: Die ermittlung physikalischer entladungsparameter in isoliergasen und isoliergasgemischen mit einer verbesserten swarm-methode. Ph.D. dissertation, Eidgenössische Technische Hochschule Zürich, Zurich, Germany, from (8) (1985) (in German)
16. Christophorou, L.G., Van Brunt, R.J.: SF_6/N_2mixtures: basic and HV insulation properties. IEEE Trans. Dielectrics and Elec. Insulation 2(5), 952–1003 (1995)