A STUDY ON CF_3I-Ar AND CF_3I-Kr MIXTURE GASES SUBSTITUTING SF_6 IN HIGH VOLTAGE EQUIPMENTS

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Abstract - The present binary mixtures of the SF_6 gas with Ar and Kr gases have not been used in many industries as long-term measures for totally eliminating the potential contribution of SF_6 to global warming. In order to gain more insight into electron transport coefficients in mixture gases as substitutes for SF_6 in high voltage equipment, transport coefficients such as electron drift velocity, density-normalized longitudinal diffusion coefficient, ratio of the longitudinal diffusion coefficient to the electron mobility, Townsend first ionization coefficient, electron attachment coefficient, and density-normalized effective ionization coefficient in CF_3I-Ar and CF_3I-Kr mixture gases are calculated and analyzed in the wide E/N range of 0.01 – 1000 Td using a two-term approximation of the Boltzmann equation for the energy. These calculated coefficients are analyzed and compared to those in pure SF_6 gas. The limiting field strength values of E/N, $(E/N)_{lim}$, of these mixture gases are also derived and compared with those of the pure SF_6 gas at different percentages of CF_3I and SF_6. The mixture gases of 70% CF_3I with Ar and Kr have $(E/N)_{lim}$ values greater than those of the pure SF_6 gas. Therefore, these mixture gases could be considered to substitute SF_6 gas in high voltage equipment.

Key words - Trifluoroiodomethane; CF_3I; SF_6; Boltzmann equation analysis; electron transport coefficients; gas mixture

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 the utilizations of those gases in the many industries [1]. In recent decades, the conventional gases such as N_2, CO_2, and air and the rare gases such as Ar, Kr, Xe, He, and Ne have been considered to mix with the SF_6 gas as a potential to reach those attempts [2]. However, the present binary mixtures of the SF_6 gas with other gases have not been used in many industries as long-term measures for totally eliminating the potential contribution of SF_6 to global warming [2].

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 [3]-[5]. 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 inertness and for fire-fighting [3]. The boiling point of CF_3I gas is higher than that of the SF_6 gas [4]. At an absolute pressure of 0.5 MPa, CF_3I becomes liquids at about 26^0C, whereas the SF_6 gas becomes liquids at about -30^0C [4]. On the other hand, the SF_6 gas is used in gas circuit breakers at 0.5 to 0.6 MPa. Therefore, it is impossible to use CF_3I gas if this gas is used at this pressure level [4]. However, in order to reduce the liquefaction temperature of CF_3I gas, Taki *et al.* [4] 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 26^0C (pure CF_3I) to about -12^0C at 0.5 MPa by using a 30% CF_3I-CO_2 mixture [5]. Therefore, it is necessary to mix the CF_3I gas with different buffer gases.

Moreover, the sets of electron collision cross sections and electron transport coefficients for atoms, molecules, and binary mixture gases are necessary for quantitative understanding of plasma phenomena. Some gases, such as rare gases (Ar, Kr, Xe, Ne, and He), N_2, CO_2, air, and O_2 mixed with each of F_2, Cl_2, and SF_6, are also necessary for many applications, such as rare-gas halide laser, plasma etching, and gaseous dielectric materials [2]. On the other hand, the collision processes and electron transport coefficients of the binary mixtures of CF_3I gas with other gases have been scarce so far. To the best of our knowledge, neither measurements nor calculations of the electron transport coefficients in the binary mixtures of the CF_3I gas with the Kr gas with the entire CF_3I concentration range have been performed previously.

In the present study, in order to gain more insight into the electron transport coefficients, the electron transport coefficients (electron drift velocity, density-normalized longitudinal coefficient, and density-normalized effective ionization coefficient) in the E/N range(ratio of the electric field E to the neutral number density N) of 10 - 1000 Td and the limiting field strength of E/N, $(E/N)_{lim}$, for the CF_3I-Ar and CF_3I-Kr mixtures are calculated by a two-term approximation of the Boltzmann equation for the energy. The negative differential conductivity (NDC) phenomena, that is, decreasing electron drift velocity with increasing electric field strength, in these binary gas mixtures are suggested. The electron transport coefficients calculated are also compared with those of pure SF_6 gas and the $(E/N)_{lim}$ values in those mixtures are also compared respectively with those of SF_6 mixtures with correlative gases (Ar and Kr) in the experiments. The binary mixtures of CF_3I gas with Ar and Kr gases with CF_3I concentration equal to about 65 - 75%, are considered for use in high voltage and many industries.

2. Calculation method of electron transport coefficients in CF_3I-Ar and CF_3I-Kr mixtures

The electron transport coefficients are calculated by sets of electron collision cross sections for gases and a two-term approximation of the Boltzmann equation for the

energy given by Tagashira *et al.* [6]. The accurate electron collision cross section sets for each gas in mixture are chosen for calculation to obtain the reliable electron transport coefficients. The electron energy distribution function (EEDF) can be computed by solving the Boltzmann equation. In this study, a two-term approximation is applied as successfully used in our previous article [7]. Based on the EEDF, f(ε, E/N), the electron drift velocity, W, the density-normalized longitudinal diffusion coefficient, ND_L, the Townsend first ionization, α, and the electron attachment coefficient, η, can be calculated as following equations:

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

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.

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

where V_1 is the speed of electron, q_T is the total cross section. 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

$$V_1 = \left(\frac{2e}{m}\right)^{1/2}; \quad \varpi_0 = V_1 N\int_0^\infty \varepsilon^{1/2} q_i F_0 d\varepsilon;$$

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

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

where q_i is the ionization cross section.

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

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

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

where $q_a(\varepsilon)$ is the attachment cross section.

The electron collision cross sections for CF_3I determined by Kimura and Nakamura [8], Ar determined by Nakamura and Kurachi [9], and Kr determined by Hayashi [10] are used throughout the present study. The set of electron collision cross sections for the CF_3I molecule [8] includes one momentum transfer, one attachment, three vibrational excitations (threshold energies of 0.032 - 0.134 eV), five electronic excitations (threshold energies of 4.7 - 9.6 eV), and one total ionization (threshold energy of 10.2 eV) cross sections.

The set of electron collision cross sections for Ar atom [9] includes one momentum transfer, five electronic excitations (threshold energies of 11.6 - 13.9 eV), and one total ionization (threshold energy of 15.69 eV) cross sections. The set of electron collision cross sections for Kr atom [10] includes one momentum transfer, fourteen electronic excitations (threshold energies of 9.915 - 13.437 eV), and one total ionization (threshold energy of 14 eV) cross sections. The accuracy of the electron collision cross section set for each gas is confirmed to be consistent with all electron transport coefficients in each pure gas.

3. Results and discussions

The results for the electron drift velocities, W, as functions of E/N for the binary mixtures of CF_3I gas with Ar and Kr gases calculated in the E/N range 10 < E/N < 1000 Td by a two-term approximation of the Boltzmann equation are shown in Figures 1-2, respectively. 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 Ar and Kr atoms, and the CF_3I molecule. These suggestions are analyzed and explained thoroughly by Chiflikian [11]. In the binary mixtures of the CF_3I gas with the Ar and Kr gases, 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 [12] for the pure SF_6 gas is shown in Figures 1-2. The calculated electron drift velocities in 70% CF_3I-Ar in the E/N ranges of E/N < 600 Td are very close to those of the pure SF_6 gas.

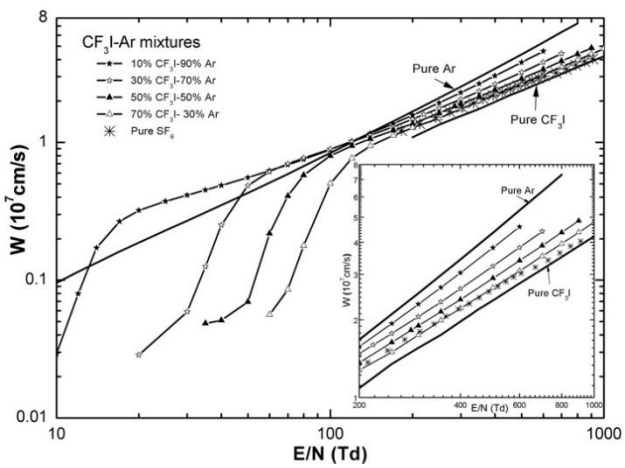

Figure 1. *Electron drift velocity, W, as functions of E/N for the CF_3I-Ar mixtures with 10%, 30%, 50%, and 70% CF_3I. The solid line and symbols show present W values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-Ar mixtures. The solid curves show present W values calculated for the pure CF_3I molecule and pure Ar atom. The star symbol shows the measurement value of the pure SF_6 [12]. The inset figure shows these results calculated in the E/N range of 200 - 1000 Td*

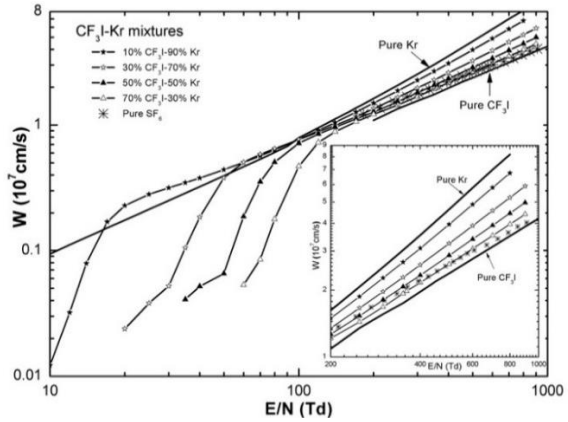

Figure 2. *Electron drift velocity, W, as functions of E/N for the CF_3I-Kr mixtures with 10%, 30%, 50%, and 70% CF_3I. The solid line and symbols show present W values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-Kr mixtures. The solid curves show present W values calculated for the pure CF_3I molecule and pure Kr atom. The star symbol shows the measurement value of the pure SF_6 [12]. The inset figure shows these results calculated in the E/N range of 200 - 1000 Td*

The results for the density-normalized longitudinal coefficients, ND_L, as functions of E/N for the binary mixtures of CF_3I gas with Ar and Kr gases calculated in the E/N range 10 < E/N < 1000 Td by a two-term approximation of the Boltzmann equation are shown in Figures 3-4, respectively.

For each E/N value, the ND_L values of the binary mixtures of the CF_3I gas with Ar and Kr gases decrease with the increase in the CF_3Icontent in the mixture. This behavior is due to the growing influence of the electron-CF_3I interaction as the CF_3Icontent increases. In these figures, on the other hand, these ND_L curves have minima in the E/N range of 15 - 170 Td for these binary mixtures. The same process responsible for the NDC region in the electron drift velocity curves in these binary mixtures caused the occurrence of these minima. Urquijo *et al.* [13] also observed the similar behavior for the C_2F_6-Ar mixtures. The density-normalized longitudinal coefficient for the pure SF_6 obtained by Aschwanden [12] is also shown in Figures 3-4 for the sake of comparison. The ND_L values of the pure SF_6 are greater than those of these binary mixtures.

The results for the density-normalized effective ionization coefficients, $(\alpha - \eta)/N$, as functions of E/N for the binary mixtures of CF_3I gas with Ar and Kr gases calculated by a two-term approximation of the Boltzmann equation are shown in Figures 5-6, respectively. In the binary mixtures of the CF_3I with the Ar and Kr gases, the values of $(\alpha - \eta)/N$ are also suggested to be between those of the pure gases, respectively. For the sake of comparison, the density-normalized effective ionization coefficient obtained by Aschwanden [12] for the pure SF_6 gas is also shown in Figures 5-6. The $(\alpha - \eta)/N$ values for 70% CF_3I mixtures with the Ar and Kr gases are very close to those of the pure SF_6 gas over E/N < 450 Td and E/N < 470 Td, respectively.

Because of the accuracy of the electron collision cross sections for the present gases and the validity of the Boltzmann equation, the present calculated results are reliable. 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.

The limiting field strength values of E/N, $(E/N)_{lim}$, at which $\alpha = \eta$ for the binary mixtures of CF_3I gas with Ar and Kr gases are derived at 133.322 Pa and shown in Figure 7. These values are also compared respectively with those of the binary mixtures of the SF_6 gas with the Ar [14] and Kr [15] gases shown in Figure 7. The $(E/N)_{lim}$ value calculated for the pure CF_3I gas is equal to 437 Td greater than the $(E/N)_{lim}$ of the pure SF_6 gas (361 Td) [12]. It can be considered as a prospective substitute for the SF_6 gas. In Figure 7, the CF_3I concentration in the binary mixtures of CF_3I gas with Ar and Kr gases equal to about 65 - 75%, is considered for use in high voltage and many industries if other chemical, physical, electrical, thermal, and economical studies are considered thoroughly.

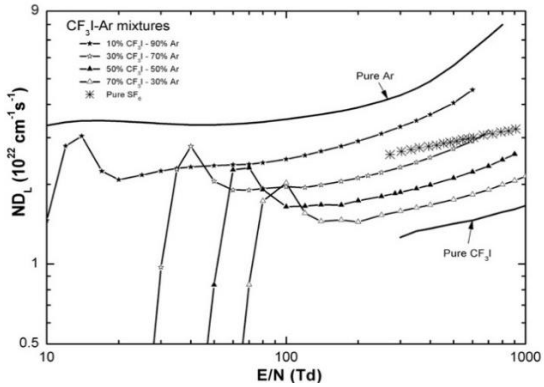

Figure 3. *Density-normalized longitudinal coefficient, ND_L, as functions of E/N for the CF_3I-Ar mixtures with 10%, 30%, 50%, and 70% CF_3I. The solid line and symbols show present ND_L values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-Ar mixtures. The solid curves show present ND_L values calculated for the pure CF_3I molecule and pure Ar atom. The star symbol shows the measurement value of the pure SF_6 [12]*

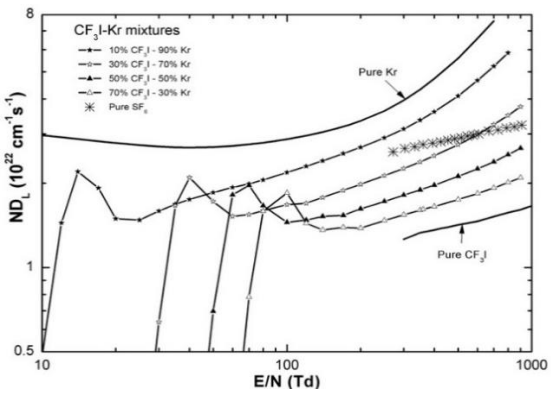

Figure 4. *Density-normalized longitudinal coefficient, ND_L, as functions of E/N for the CF_3I-Kr mixtures with 10%, 30%, 50%, and 70% CF_3I. The solid line and symbols show present ND_L values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-Kr mixtures. The solid curves show present ND_L values calculated for the pure CF_3I molecule and pure Kr atom. The star symbol shows the measurement value of the pure SF_6 [12]*

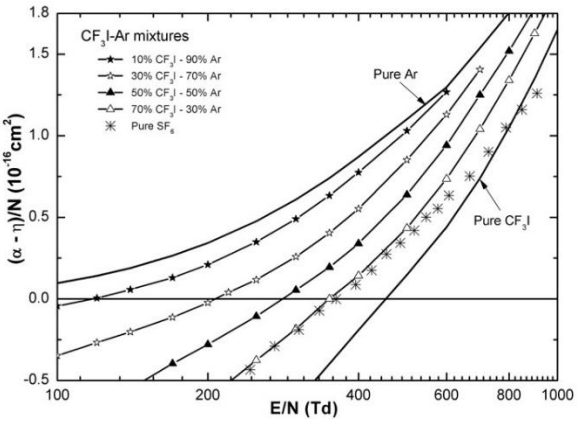

Figure 5. *Density normalized effective ionization coefficient, (α - η)/N, as functions of E/N for the CF_3I-Ar mixtures with 10%, 30%, 50%, and 70% CF_3I. The solid line and symbols show present (α - η)/N values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-Ar mixtures. The solid curves show present (α - η)/N values calculated for the pure CF_3I molecule and pure Ar atom. The star symbol shows the measurement value of the pure SF_6 [12]*

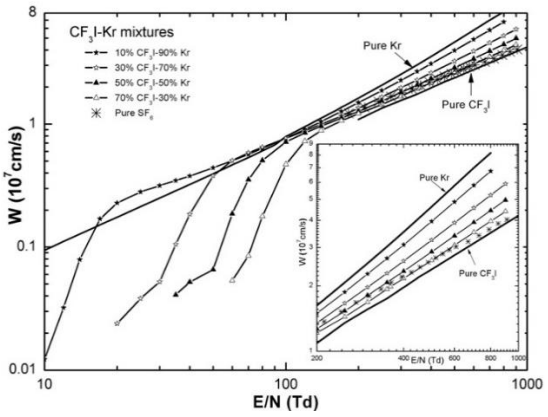

Figure 6. *Density normalized effective ionization coefficient, (α - η)/N, as functions of E/N for the CF_3I-Kr mixtures with 10%, 30%, 50%, and 70% CF_3I. The solid line and symbols show present (α - η)/N values calculated using a two-term approximation of the Boltzmann equation for the CF_3I-Kr mixtures. The solid curves show present (α - η)/N values calculated for the pure CF_3I molecule and pure Kr atom. The star symbol shows the measurement value of the pure SF_6 [12]*

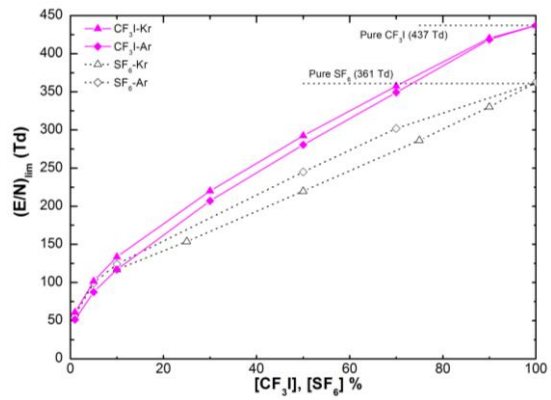

Figure 7. *Limiting field strength values of E/N, $(E/N)_{lim}$, as functions of the percentage of CF_3I gas for the binary mixtures CF_3I-Ar and CF_3I-Kr. The solid line and solid symbols show present $(E/N)_{lim}$ values for these binary mixtures calculated using a two-term approximation of the Boltzmann equation. The dotted curves and the open symbols show $(E/N)_{lim}$ values for the binary mixtures SF_6-Ar [14] and SF_6-Kr [15]*

4. Conclusion

The electron drift velocity, density-normalized longitudinal coefficient, and density-normalized effective ionization coefficient in the binary mixtures in CF_3I with Ar and Kr gases are calculated using a two-term approximation of the Boltzmann equation for the energy in the E/N range of 10 - 1000 Td for the first time. The NDC phenomena in these binary gas mixtures are suggested. The electron transport calculated coefficients are also compared with those of the pure SF_6 gas in experiments. The limiting field strength values of E/N for the binary mixtures of 70% CF_3I gas with Ar and Kr gases are determined and greater than those of the pure SF_6 gas. Therefore, these binary mixtures with CF_3I concentration equal to about 65 - 75% are considered for 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.

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