



Ionization and Attachment Coefficients in C₄F₇N Gas Measured by the Steady-State Townsend Method

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Abstract: The normalized Townsend first ionization coefficient α/N and normalized attachment coefficient η/N in pure C₄F₇N were measured by using the steady-state Townsend (SST) method for a range of reduced electric fields E/N from 750 to 1150 Td at room temperature (20 °C). Meanwhile, the effective ionization coefficients are obtained. All SST experimental results show good agreement with pulsed Townsend (PT) experiment results. Comparisons of the critical electric fields of C₄F₇N with SF₆ and other alternative gases such as c-C₄F₈ and CF₃I indicate that C₄F₇N has a better insulation performance with a much higher normalized critical electric field at 959.19 Td.

Keywords: C_4F_7N ; ionization coefficient; attachment coefficient; critical electric field; steady-state Townsend method; ionization; plasma; gas discharge; high voltage; SF_6

1. Introduction

As is well known, sulfur hexafluoride (SF₆), with a good insulation performance and thermal stability, is widely used as an insulation gas and arc quenching medium in many fields such as high-voltage engineering and electrical power applications [1,2]. Unfortunately, as one of the six greenhouse gases, SF₆ has an extremely high global warming potential (GWP₁₀₀) at 23,500 times that of CO₂ [3]. Thus, in order to slow down global warming, searching for an environment-friendly insulation gas to reduce the use of SF₆ is of great importance.

During the last decades, there has been some research about the discharge characteristics of $c-C_4F_8$ [4–6] and CF_3I [7–9], which have been thought of as potential alternative gases to SF₆. Recently, as another newly potential substitutional insulation gas of SF₆, fluoronitriles (C₄F₇N, also known as 3M Novec-4710), which have a GWP value of 2100, have been designed and have attracted great attention [10]. Although C₄F₇N gas has a good insulation performance, because of its high boiling temperature, it has to be mixed with other buffer gases such as CO₂ to adapt to the operating conditions under high gas pressure [11]. Nevertheless, it is worthy to obtain the basic electron swarms of C₄F₇N, such as the ionization coefficient α and attachment coefficient η , which can be used to evaluate the insulation performance of gases [12].



However, it should be noted that there has been less investigation on the ionization coefficients and attachment coefficients in C_4F_7N . It has been reported once that the normalized effective ionization coefficient ($\alpha - \eta$)/N within a small range of reduced electric fields (per gas density N) *E*/N was obtained with the steady-state Townsend (SST) method by Nechmi et al. [13]. The electron rate and transport coefficients in pure C_4F_7N were obtained with the pulsed Townsend (PT) method by Chachereau et al. [14]. In the present work, both normalized ionization and normalized attachment coefficients, as well as effective ionization coefficients of C_4F_7N , were measured by the SST method, a kind of method which has been widely used to measure these coefficients in different gases like SF₆ [15–17], c-C₄F₈ [4], CF₃I [8], and so on. It should also be noticed that although the SST method has been well developed during the last decades, and the innovation of the method itself may not be high, the experiments using this method to measure and obtain the discharge parameters and evaluate the insulation abilities of new insulation gases are still significant.

2. Experiments

The experiments were carried out on an SST apparatus setup in Wuhan University, which the details have been described in previous work [18,19]. The reliability of this SST apparatus has been verified by experiments on known gases such as SF₆ and N₂, which showed this SST apparatus has great measurement accuracy (<2%) [19]. The theoretical equations used to obtain the experimental results have been introduced as well. Through the update of vacuum-tight gaskets, the background vacuum degree in the ionization chamber can reach ~1.0 × 10⁻⁵ Pa at room temperature with a leak rate less than 0.10 Pa/h, which can minimize the effect of the pressure change on experimental results. In the present study, the range of reduced electric fields *E/N* applied for the SST experiment varied from 750 to 1150 Td (1 Td = 10^{-21} Vm²), since the insulation performance of C₄F₇N is much better than SF₆ as reported [13,14]. The purity of C₄F₇N gas (produced by Beijing Yuji Science and Technology Co., Ltd., China) used in the present study was more than 99.6%, and the impurities were mainly air (~0.3%). The gas pressure was 500 Pa at 20 °C. The initial electron current *I*₀ of the electron avalanche was about 4 pA, and current *I* in the nonself-sustained discharge stage was also on the order of picoampere (pA).

3. Results and Discussion

3.1. Ionization and Attachment Coefficients

The normalized ionization coefficient α/N and the attachment coefficient η/N of C_4F_7N gas for 750 Td < E/N < 1150 Td were measured respectively. The results were compared to that of SF₆ gas and shown in Figure 1. It was found that with an increasing E/N, the α/N of C_4F_7N showed a growing trend, while the η/N showed a clear decreasing trend, which were similar to that of SF₆ [3]. Meanwhile, compared to SF₆ gas, it was apparent that the values of η/N in C_4F_7N were much higher than that of SF₆ for the same E/N, which could be explained with the attachment cross-sections in different gases. The higher the attachment cross section is, the stronger the ability of electron attachment is. The total attachment cross-sections of C_4F_7N [14], SF₆ [9], and other gases [9,20] have been compared and plotted in Figure 2. As reported [14], the total attachment cross-section in C_4F_7N was larger than that of SF₆ above 0.1 eV, while the values of α/N were a little bit smaller than that of SF₆. Therefore, for C_4F_7N and SF₆ with similar values of ionization coefficients, C_4F_7N had a better insulation performance due to the much higher attachment coefficient than that of SF₆.



Figure 1. Values of α/N and η/N as a function of E/N in C₄F₇N gas in the present work and their comparison to SF₆ gas.



Figure 2. Comparison of total attachment cross-sections of C₄F₇N, SF₆, c-C₄F₈, and CF₃I.

The α/N and η/N measured by the SST method in this work have also been compared to the PT method measured by Chachereau et al. [14], which are plotted in Figure 3. It should be noted that the values of α/N and η/N used for comparisons with the PT method were not given directly in Chachereau's work; they were calculated from parameters such as electron drift velocity (w_e), ionization rate coefficient (k_i), and attachment rate coefficient (k_a). It can be found that the trends of varying α/N and η/N in this work showed good agreement with PT experiments. Significantly, the fluctuation of PT was much larger than that of SST, which may be caused by the different experimental principles and conditions, such as a much lower pressure (100 Pa), in the PT experiment.

3.2. Effective Ionization Coefficients

The normalized effective ionization coefficient $(\alpha - \eta)/N$ of C_4F_7N , in a range of E/N from 750 to 1150 Td, at 20 °C was obtained as well. Figure 4 presents the value $(\alpha - \eta)/N$ as a function of E/N in C_4F_7N , and its comparisons with SF_6 [3], c- C_4F_8 [4], CF_3I [7], C_4F_7N [13,14], and C_4F_7N/N_2 mixtures [18] are reported. Notably, the E/N of C_4F_7N was much greater than other kinds of gases for the same $(\alpha - \eta)/N$, which suggests that the insulation ability of C_4F_7N is much stronger. Once the C_4F_7N gas mixed with buffer gas N_2 , the insulation performances of the mixtures were much weaker than pure C_4F_7N , since N_2 is an electrically neutral gas whose attachment coefficients is 0. Meanwhile,

the variety of $(\alpha - \eta)/N$ with E/N showed a linear trend for all these gases nearby the normalized critical electric field $(E/N)_{lim}$ (for $\alpha - \eta = 0$). Moreover, compared to the data reported, our results of normalized effective coefficients in C₄F₇N were in good agreement with that of Nechmi et al. [13] as well as Chachereau [14]. Meanwhile, more values in the E/N that varied from 750 to 1150 Td were obtained in the present work.



Figure 3. (a) Comparison of values of α/N measured with the steady-state Townsend (SST) method in this work and the pulsed Townsend (PT) method [14]; (b) Comparison of values of η/N measured with SST method in this work and PT method [14].

According to the bond length values of the C_4F_7N molecule [21], the structure of the C_4F_7N molecule could be drawn as in Figure 5. A recent study [22] shows that the bonds of C-1 to C-2 and C-1 to C-3 have the smallest bond energy, which is 3.812 eV/atom. The second smallest bond energy is 4.556 eV/atom, which belongs from C-1 to F-1. The bond energy of S-F in SF₆ is 3.432 eV/atom, which is smaller than the smallest bond energy in C_4F_7N . It is well known that the smaller the bond energy is, the weaker the interaction between atoms will be. Since the C_4F_7N and SF_6 molecules have a strong ability to attach electrons, they can easily adsorb electrons and, hence, become negatively charged molecules. Then, these charged molecules could accelerate under an electric field, applied between two plate electrodes, and collide with other molecules. Thus, the collision may lead to the breaking of weak bonds and forming new particles such as F in SF₆ and CF₃ in C₄F₇N. For the same electric field, the bonds of SF₆ are easier to be broken than bonds C-1 to C-2 (or C-3) of C_4F_7N . Consequently, the new particles formed in the SF_6 gas, such as F, would further take part in the discharging process. However, the CF₃ formed in C₄F₇N more easily adorbs electrons than the F formed in SF₆, since there is more F in CF_3 , and fluorine has strong electronegativity. Then, SF_6 is more likely to exhibit ionization characteristics, which could lead to the higher effective ionization coefficients of SF_6 than that of C_4F_7N for the same E/N.



Figure 4. Comparison of the normalized effective ionization coefficient in different gases.



Figure 5. The molecule structure of C_4F_7N gas.

3.3. Critical Electric Fields

In order to more clearly compare and evaluate the insulation performance quantitatively, the $(E/N)_{lim}$ of these four gases have been sorted out in Table 1. It is apparent that C_4F_7N has a superior performance of gas insulation, as it had a high $(E/N)_{lim}$ value (959.19 Td). The relative deviation of $(E/N)_{lim}$ of C_4F_7N measured in this work was about 2.41% of that measured by Nechmi et al. [13] and was about 1.73% of that measured by Chachereau et al. [14], which could testify that the experiments in the present study were of high credibility at the same time. Furthermore, it could be found that the value of $(E/N)_{lim}$ of C_4F_7N was about 2.68 times that of pure SF₆ gas. However, the dielectric strength of C_4F_7N at atmospheric pressure was about 2 times that of SF₆ as reported [10], slightly less than 2.68 times, which could be caused by different behaviors of ion kinetics under different gas pressures [14]. The higher the gas pressure, the greater the effect of ion kinetics.

Table 1. The values of $(E/N)_{lim}$ in different gases.

Gas Type	(E/N) _{lim} /Td
C ₄ F ₇ N	959.19 [present work]
	981.84 [13]
	975 [14]
c-C ₄ F ₈	408.68 [4]
CF ₃ I	440.19 [7]
SF ₆	355.27 [13]
	358.66 [14]
	351.80 [16]

4. Conclusions

In the present work, both the α/N and η/N in C_4F_7N have been measured by using the SST method for E/N from 750 to 1150 Td at 20 °C, and they were compared to that of pure SF₆ gas. Moreover, the value of $(\alpha - \eta)/N$ was obtained for 750 Td < E/N < 1150 Td. All results measured by the SST method showed good agreement with PT experiments. The critical electric field $(E/N)_{lim}$ of C_4F_7N was about 959.19 Td. The comparison indicated that the insulation performance of C_4F_7N is superior to that of SF₆, and C_4F_7N could be considered as a candidate insulation gas to replace SF₆ in the high-voltage engineering field.

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References

- Christophorou, L.G.; Van Brunt, R.J. SF₆/N₂ Mixtures Basic and HV Insulation Properties. *IEEE Trans. Dielectr. Electr. Insul.* 1995, 2, 952–1003. [CrossRef]
- Dincer, M.S.; Raju, G.G. Ionization and Attachment Coefficients in SF₆+N₂ Mixtures. *IEEE Trans. Electr. Insul.* 1984, *EI*-19, 40–44. [CrossRef]
- 3. Christophorou, L.G.; Olthoff, J.K. Electron Interactions with SF₆. J. Phys. Chem. Ref. Data 2000, 29, 267–330. [CrossRef]
- 4. Yamaji, M.; Nakamura, Y.; Morokuma, Y. Measurements of ionization and attachment coefficients in 0.468% and 4.910% c-_CC₄F₈/Ar mixtures and pure c-C₄F₈. *J. Phys. D Appl. Phys.* **2004**, *37*, 432–437. [CrossRef]
- 5. de Urquijo, J.; Basurto, E. Electron attachment, ionization and drift in c-C₄F₈. *J. Phys. D Appl. Phys.* **2001**, *34*, 1352–1354. [CrossRef]
- Naidu, M.S.; Prasad, A.N.; Craggs, J.D. Electron transport, attachment and ionization in c-C₄F₈ and iso-C₄F₈. *J. Phys. D Appl. Phys.* **1972**, *5*, 741–746. [CrossRef]
- Li, X.; Zhao, H.; Wu, J.; Jia, S. Analysis of the insulation characteristics of CF₃I mixtures with CF₄, CO₂, N₂, O₂ and air. J. Phys. D Appl. Phys. 2013, 46, 345203–345210. [CrossRef]
- Hasegawa, H.; Date, H.; Shimozuma, M.; Itoh, H. Properties of Electron Swarms in CF₃I. *Appl. Phys. Lett.* 2009, *95*, 101504–101506. [CrossRef]
- 9. Raju, G.G. *Gaseous Electronics: Tables, Atoms, and Molecules;* CRC Press Taylor & Francis Group: Boca Raton, FL, USA, 2012.
- Kieffel, Y.; Irwin, T.; Ponchon, P.; Owens, J. Green Gas to Replace SF₆ in Electrical Grids. *IEEE Power Energy* Mag. 2016, 14, 32–39. [CrossRef]
- Kieffel, Y.; Biquez, F.; Poncho, P. Alternative gas to SF₆ for use in high voltage switchgears: g³. In Proceedings of the 23rd International Conference on Electricity Distribution (CIRED), Lyon, France, 15–18 June 2015; p. 230.
- 12. Townsend, J.S. The Conductivity produced in Gases by the Motion of Negatively-charged Ions. *Nature* **1900**, 62, 340–341. [CrossRef]
- 13. Nechmi, H.E.; Beroual, A.; Giodet, A.; Vinson, P. Effective Ionization Coefficient and Limiting Field Strength of Fluoronitriles-CO₂ Mixtures. *IEEE Trans. Dielectr. Electr. Insul.* **2017**, 24, 886–892. [CrossRef]
- Chachereau, A.; Hösl, A.; Franck, C.M. Electrical insulation properties of the perfluoronitrile C₄F₇N. *J. Phys.* D Appl. Phys. 2018, 51, 495201–495210. [CrossRef]

- 15. Bhalla, M.S.; Craggs, J.D. Measurement of Ionization and Attachment Coefficients in Sulphur Hexafluoride in Uniform Fields. *Proc. Phys. Soc.* **1962**, *80*, 151–160. [CrossRef]
- 16. Raju, G.G.; Dincer, M.S. Measurement of ionization and attachment coefficients in SF₆ and SF₆+N₂. *J. Appl. Phys.* **1982**, *53*, 8562–8567. [CrossRef]
- 17. Fréchette, M.F. Experimental study of SF₆/N₂ and SF₆/CCl₂F₂ mixtures by the steady-state Townsend method. *J. Appl. Phys.* **1986**, *59*, 3684–3693. [CrossRef]
- Long, Y.; Guo, L.; Shen, Z.; Chen, C.; Chen, Y.; Li, F. Ionization and Attachment Coefficients in C₄F₇N/N₂ Gas Mixtures for Use as a Replacement to SF₆. *IEEE Trans. Dielectr. Electr. Insul.* 2019, 26, 1358–1362. [CrossRef]
- Long, Y.; Guo, L.; Shen, Z.; Chen, C.; Zhou, W.; Liu, W. Investigation of Ionization Characteristics of C₄F₇N-N₂ Gas Mixtures as Alternative Gas to SF₆ by Steady-state Townsend Method. *High Volt. Eng.* 2019, 45, 1064–1070.
- 20. Christophorou, L.G.; Olthoff, J.K. Electron Interactions with CF₆I. *J. Phys. Chem. Ref. Data* **2000**, *29*, 553–569. [CrossRef]
- Li, Y.; Zhang, X.; Xiao, S.; Chen, Q.; Tang, J.; Chen, D.; Wang, D. Decomposition Properties of C₄F₇N/N₂ Gas Mixture: An Environmentally Friendly Gas to Replace SF₆. *Ind. Eng. Chem. Res.* 2018, 57, 5173–5182. [CrossRef]
- 22. Wang, C.; Wu, Y.; Sun, H.; Duan, J.; Niu, C.; Yang, F. Thermophysical Properties Calculation of C₄F₇N/CO₂ mixture Based on Computational Chemistry—A Theoretical Study of SF₆ Alternative. In Proceedings of the 4th International Conference on Electric Power Equipment-Switching Technology (ICEPE-ST), Xi'an, China, 22–25 October 2017; pp. 255–258.



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