Measurement of Ionization and Attachment Coefficients in C$_4$F$_7$N/CO$_2$ Gas Mixture as Substitute Gas to SF$_6$

YUNXIANG LONG$^{1,2}$, LIPing GUO$^{1,2}$, CHENG CHEN$^{1,2}$, ZHENYU SHEN$^{1,2}$, YIHENG CHEN$^{1,2}$, FANG LI$^{1,2}$, AND WENJUN ZHOU$^3$, (Senior Member, IEEE)

$^1$Hubei Key Laboratory of Nuclear Solid Physics, Key Laboratory of Artificial Micro- and Nano-Structures of Ministry of Education, Wuhan University, Wuhan 430072, China
$^2$School of Physics and Technology, Wuhan University, Wuhan 430072, China
$^3$School of Electrical Engineering and Automation, Wuhan University, Wuhan 430072, China

Corresponding author: Liping Guo (guolp@whu.edu.cn)

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ABSTRACT As one of the alternative insulation gases, C$_4$F$_7$N/CO$_2$ gas mixtures have attracted much attention recently. In this study, the normalized Townsend first ionization coefficient $\alpha/N$ and the normalized attachment coefficient $\eta/N$ were measured in C$_4$F$_7$N/CO$_2$ gas mixtures by using the Steady-State Townsend (SST) method over a range of electric fields $E/N$ from 100 to 550 Td. The concentrations of C$_4$F$_7$N studied were 4.99, 7.00, 9.00, 13.13 and 20.02%, and the gas pressure were 500 Pa at 20°C. The effective ionization coefficients and the critical electric field were obtained at 500, 1000 and 2000 Pa to investigate the effect of gas pressure on dielectric strength in 9.00% C$_4$F$_7$N/91.00% CO$_2$ respectively. The results indicated that the $(E/N)_{\text{lim}}$ have not significant change on the SST experimental conditions. Meanwhile, a comparison with SF$_6$ gas indicates that 20.02% C$_4$F$_7$N/79.98% CO$_2$ has a similar insulation performance with $(E/N)_{\text{lim}}$ as 358.82 Td. However, when the liquefaction temperature is considered, the C$_4$F$_7$N/CO$_2$ gas mixtures with 9.00% C$_4$F$_7$N could be considered as an appropriate substitute gas for SF$_6$, because the critical electric field of this gas mixture is over 70% of SF$_6$ gas and could be used under -15°C at 0.7 MPa.

INDEX TERMS C$_4$F$_7$N/CO$_2$, ionization coefficient, attachment coefficient, critical electric field, steady-state Townsend method.

I. INTRODUCTION

Due to the great insulation ability, toxic safety and the heat transfer properties, sulfur hexafluoride (SF$_6$) is widely used in high voltage applications, such as gas insulated switchgear (GIS), gas insulated transmission line (GIL) and so on [1]–[3]. However, as one of the six restricted greenhouse gases listed in the Kyoto Protocol in 1997, SF$_6$ has an extremely high value of global warming potential (GWP$_{100}$), which is 22800 times that of CO$_2$ [4], [5]. Therefore, the search for an environment-friendly insulation gas to reduce even replace the SF$_6$ in power systems has reached the urgent stage.

Recently, as a newly potential candidate gas, Fluoronitrile (C$_4$F$_7$N), whose value of GWP is about 2100, is attracting much attention. [2], [6]–[11]. However, pure C$_4$F$_7$N have to be diluted with the buffer gas such as N$_2$, CO$_2$ or dry air for electrical applications due to its high boiling point as $-4.7^\circ$C [6]. Then, C$_4$F$_7$N/CO$_2$ gas mixtures could be considered as the alternative gases to SF$_6$. So far, most of the researches focus on the breakdown characteristics and other insulation properties at high gas pressure in C$_4$F$_7$N/CO$_2$ gas mixtures. As far as we know, there is little reports about the transport parameters in C$_4$F$_7$N/CO$_2$ mixtures. Nechmi et al. [7] used steady-state Townsend (SST) method to obtained the normalized effective ionization coefficient $(\alpha-\eta)/N$ (per unit gas density $N$) of C$_4$F$_7$N/CO$_2$ gas mixture with the concentration of C$_4$F$_7$N as 3.7, 6.7 and 20 %, within a small range of reduced electric fields $E/N$ as 200 $\sim$ 430 Td.
Chachereau et al. [10], [11] used Puled Townsend (PT) method to obtained the electron rate and transport coefficients such as effective ionization rate coefficient and electron drift velocity in C$_4$F$_7$N/CO$_2$ mixtures with the concentration of C$_4$F$_7$N varying from 1% to 40%. Nevertheless, there is few reports about the normalized ionization coefficient $\alpha/N$ and the normalized attachment coefficient $\eta/N$ measured by using the SST method in C$_4$F$_7$N/CO$_2$ with different ratio of C$_4$F$_7$N.

In present work, the normalized ionization coefficients $\alpha/N$, the normalized attachment coefficients $\eta/N$, the normalized effective ionization coefficient $(\alpha - \eta)/N$ were measured by using the SST method. The critical electric fields $(E/N)_{\text{lim}}$ (for $\alpha - \eta = 0$) of C$_4$F$_7$N/CO$_2$, which can be used as references for evaluating the dielectric strength [12], were obtained as well. Moreover, in order to investigate the effect of gas pressure on the SST experimental conditions, the $(\alpha - \eta)/N$ and $(E/N)_{\text{lim}}$ were obtained at 500 Pa, 1000 Pa and 2000 Pa respectively. According to the requirements of the funding program, the C$_4$F$_7$N/CO$_2$ mixture should achieve 80% of the insulation abilities of SF$_6$ and could be used under -15°C. Then, the liquefaction temperature of gas mixtures with different contents of C$_4$F$_7$N were calculated according to the Antoine equation. Finally, several C$_4$F$_7$N/CO$_2$ mixtures were suggested as potential alternative gases when considering the critical electric fields and boiling points.

II. EXPERIMENTAL

The measurements of the C$_4$F$_7$N/CO$_2$ mixtures were performed on the SST apparatus setup in Wuhan University [13], [14]. The accuracy of the measurement is less than 2%. All equipment of the SST experimental platform have been verified by the Hubei Institute of Metrology and Testing Technology. As described in the previous work [13], the apparatus consist 5 parts, and the one of the important parts is the vacuum system. The background vacuum degree in the ionization chamber can reach approximately $1.0 \times 10^{-5}$ Pa at 20°C. The low leak rate of pressure as 0.07 Pa/h guarantee the stable gas pressure as 500 ~ 2000 Pa during the SST experiments. Additionally, the precision of the measurements of discharge currents is less than 1.5%. The purity of the C$_4$F$_7$N gas (produced by Beijing Yuji Science and Technology Co., Ltd.) used in this study is higher than 99.6%, and the purity of CO$_2$ gas is 99.99%. The partial pressure method is used to compound all the gas mixtures with C$_4$F$_7$N concentrations (molar fractions) of 4.99, 7.00, 9.00, 13.13 and 20.02% respectively. The gas pressure for these mixtures was 500 Pa at 20°C during the SST experiments. The reduced electric fields $(E/N)$ applied in the experiments varied from 100 to 550 Td (where 1 Td = $10^{-10}$ V m$^{-2}$).

As reported [7], [11], [15], the secondary ionization coefficient $\gamma$ can be considered to be negligible at the low gas pressure. Then, the theoretical equation, which is widely used in SST experiments, for the current $I$ of picoampere (pA) order in the non-self-sustained discharge stage could be expressed as follows,

$$I = \frac{\alpha}{\alpha - \eta} \times \exp[(\alpha - \eta) d] - \frac{\eta}{\alpha - \eta}$$  

where $I_0$ is the initial electron current, $d$ is the electrode separation distance. In this study, $I_0$ is controlled to 4 pA, and $d$ could be adjusted in the range of 0 ~ 15 mm with 1.5 mm as step size.

III. RESULTS AND DISCUSSION

A. IONIZATION AND ATTACHMENT COEFFICIENTS

The normalized ionization coefficient $\alpha/N$ and the attachment coefficient $\eta/N$ of C$_4$F$_7$N/CO$_2$ mixtures were measured for 100 Td < $(E/N) < 550$ Td, and the results with the comparison to SF$_6$ gas [3] are shown in Figures 1 and 2. It was found that with increasing $(E/N)$, the value of $\alpha/N$ in each kind of C$_4$F$_7$N/CO$_2$ mixture shows a growth trend, while $\eta/N$ shows...
In order to provide a comparison and evaluation of the insulation performance to pure SF\textsubscript{6}, the results of the normalized effective ionization coefficient (\(\alpha/\eta\)/N) for SF\textsubscript{6} [3] and CO\textsubscript{2} [16] could be found that comparing to pure CO\textsubscript{2} gas, there are negative values of (\(\alpha−\eta\)/N) when the C\textsubscript{4}F\textsubscript{7}N was added before 200 Td. Obviously, with increasing E/N, the value of (\(\alpha−\eta\)/N) increases. Moreover, for the same E/N, the (\(\alpha−\eta\)/N) values for the mixtures with higher concentrations of C\textsubscript{4}F\textsubscript{7}N gas are smaller. Significantly, over the experimental scale of E/N, the value of (\(\alpha−\eta\)/N) of 20.02%C\textsubscript{4}F\textsubscript{7}N/79.98%CO\textsubscript{2} mixture is most similar to that of SF\textsubscript{6}, which indicates that 20.02%C\textsubscript{4}F\textsubscript{7}N/79.98%CO\textsubscript{2} mixture has the similar insulation performance to pure SF\textsubscript{6} near the critical electric field (E/N)\textsubscript{lim}. Meanwhile, the other four mixtures show higher coefficients than 20.02%C\textsubscript{4}F\textsubscript{7}N/79.98%CO\textsubscript{2} and SF\textsubscript{6} gas for the same E/N. It is apparent that a higher (\(\alpha−\eta\)/N) leads to easier ionization processes, which means the insulation performance is weaker for the gas with higher (\(\alpha−\eta\)/N). Therefore, the insulation ability of C\textsubscript{4}F\textsubscript{7}N/CO\textsubscript{2} mixtures with C\textsubscript{4}F\textsubscript{7}N concentrations less than 20% is weaker than that of SF\textsubscript{6}.

### B. EFFECTIVE IONIZATION COEFFICIENT

The results of the normalized effective ionization coefficient (\(\alpha−\eta\)/N) in the C\textsubscript{4}F\textsubscript{7}N/CO\textsubscript{2} mixtures were obtained and plotted in Figure 3, which were compared with the (\(\alpha−\eta\)/N) coefficients for SF\textsubscript{6} [3] and CO\textsubscript{2} [16].

![Figure 3](image3.png)

**FIGURE 3.** Comparison of normalized effective ionization coefficient (\(\alpha−\eta\)/N) in different gases.

![Figure 4](image4.png)

**FIGURE 4.** Comparison of the values and variations of critical electric fields (E/N)\textsubscript{lim} in different gases.

| Gas Type          | Concentration of C\textsubscript{4}F\textsubscript{7}N or SF\textsubscript{6} | (E/N)\textsubscript{lim} / Td |
|-------------------|--------------------------|-----------------------------|
| C\textsubscript{4}F\textsubscript{7}N/CO\textsubscript{2} | 4.99%                     | 211.86                      |
|                   | 7.00%                     | 236.93                      |
|                   | 9.00%                     | 253.08                      |
|                   | 13.13%                    | 292.44                      |
|                   | 20.02%                    | 358.82                      |
| C\textsubscript{4}F\textsubscript{7}N/\textsubscript{N}_\textsubscript{2} \textsuperscript{[13]} | 5.07%                     | 262.53                      |
|                   | 7.00%                     | 289.58                      |
|                   | 9.00%                     | 305.93                      |
|                   | 13.10%                    | 345.06                      |
|                   | 19.09%                    | 394.72                      |
| SF\textsubscript{6} \textsuperscript{[3]}       | 99.999%                   | 356.77                      |

A downward trend obviously, which is similar to the trends of SF\textsubscript{6}. Moreover, with the concentration of C\textsubscript{4}F\textsubscript{7}N increases, the values of (\(\alpha/N\)) decrease and the values of (\(\eta/N\)) increase for the same E/N, which indicates that C\textsubscript{4}F\textsubscript{7}N is a strongly electronegative gas [6]. Moreover, it can be noted that both the values of (\(\alpha/N\)) and (\(\eta/N\)) for same E/N in the C\textsubscript{4}F\textsubscript{7}N/CO\textsubscript{2} mixtures are smaller than that of SF\textsubscript{6}.

### C. CRITICAL ELECTRIC FIELD (E/N)\textsubscript{lim}

In order to provide a comparison and evaluation of the insulation performance of these gases quantitatively, the results of critical electric fields (E/N)\textsubscript{lim} in C\textsubscript{4}F\textsubscript{7}N/CO\textsubscript{2} mixtures are obtained by linear fitting due to the linear variation of (\(\alpha−\eta\))/N with E/N near (\(\alpha−\eta\))/N = 0, and the comparisons with that of C\textsubscript{4}F\textsubscript{7}N/\textsubscript{N}_\textsubscript{2} \textsuperscript{[13]} gas mixtures as well as SF\textsubscript{6} \textsuperscript{[13]} have been sorted into Table 1 and plotted in Figure 4. The strength of (E/N)\textsubscript{lim} could indicate the insulation ability of insulation gases since when E/N is larger than (E/N)\textsubscript{lim}, the gas shows the ionization characteristics of electrons. Therefore, the higher (E/N)\textsubscript{lim} means the gas would keep attachment characteristics under higher E/N, which indicates that gas have better insulation performance. It could be found from Table 1 that the 20.02%C\textsubscript{4}F\textsubscript{7}N/79.98%CO\textsubscript{2} mixture provides a similar gas insulation performance to pure SF\textsubscript{6} gas since the value of (E/N)\textsubscript{lim} of this mixture is 358.82 Td, and the value of (E/N)\textsubscript{lim} is 356.77 Td in SF\textsubscript{6} gas \textsuperscript{[13]}. Meanwhile, it is worth noting that the value of (E/N)\textsubscript{lim} in 13.13%C\textsubscript{4}F\textsubscript{7}N/86.87%CO\textsubscript{2} could achieve approximately 80% of that in SF\textsubscript{6}, and 9.00%C\textsubscript{4}F\textsubscript{7}N/91.00%CO\textsubscript{2} could achieve more than 70% of that in SF\textsubscript{6}. Moreover, it should be noticed that there are several researches which showed the dielectric strength in 9.00%C\textsubscript{4}F\textsubscript{7}N/91.00%CO\textsubscript{2} mixtures could achieve more than 80% that of SF\textsubscript{6} over 0.2 MPa.
in power-frequency breakdown tests [17]. Thus, from the perspective of the value of \((E/N)_{\text{lim}}\), the mixtures of \(9.00\% \sim 13.00\%\) C\(_4\)F\(_7\)N in CO\(_2\) have potential for using as alternative gases to SF\(_6\).

As shown in Figure 4, there is a synergism of \((E/N)_{\text{lim}}\) in C\(_4\)F\(_7\)N/CO\(_2\) mixtures measured in this work, which also has a good agreement with the results obtained by PT method [10]. However, the results obtained in this work and Chachereau’s work [10] were lower than that reported in Nechmi’s [7] work. According to the report, the reason to this differences could be the scatter in the data measured by Nechmi. Moreover, it could be found that the comparison of \((E/N)_{\text{lim}}\) of the C\(_4\)F\(_7\)N/CO\(_2\) mixture and the C\(_4\)F\(_7\)N/N\(_2\) mixture [13] shows that C\(_4\)F\(_7\)N/N\(_2\) has a better insulation ability than C\(_4\)F\(_7\)N/CO\(_2\) with almost the same concentration of C\(_4\)F\(_7\)N, which shows similar phenomena in SF\(_6\)/N\(_2\) and SF\(_6\)/CO\(_2\) mixtures as reported in the literature [18]. According to previous studies [16], the value of effective ionization coefficients of CO\(_2\) is higher than that of N\(_2\) for the same \(E/N\), which indicates that it is easier for CO\(_2\) to show the ionization characteristics in discharge. Therefore, for the same concentration of electronegative gas like C\(_4\)F\(_7\)N, the mixtures with N\(_2\) are more stable to be ionized, which means that the mixtures with CO\(_2\) have weaker insulation performance in this situation.

**D. INFLUENCE OF GAS PRESSURE ON \((\alpha - \eta)/N\) AND \((E/N)_{\text{lim}}\) IN 9.00\% C\(_4\) F\(_7\) N/91.00\% CO\(_2\) MIXTURES**

It was reported that the breakdown voltage of C\(_4\)F\(_7\)N/CO\(_2\) gas mixture is higher than that of C\(_4\)F\(_7\)N/N\(_2\) gas mixture at high pressure [19], which is opposite to the SST experimental results mentioned above. Meanwhile, the electric strength in C\(_4\)F\(_7\)N/CO\(_2\) gas mixture is higher than the \((E/N)_{\text{lim}}\) measure by SST method. This interesting phenomenon were found by Chachereau [10], [11] with PT experiments as well, and the different gas pressure used in these experiments were considered as an important factor to effect the result.

In order to investigate the influence of gas pressure on \((\alpha - \eta)/N\) and \((E/N)_{\text{lim}}\) at low pressure in SST experiments, the 9.00\%C\(_4\)F\(_7\)N/91.00\%CO\(_2\) were chosen and the gases pressure were chosen as 500, 1000 and 2000 Pa respectively in this work. As reported, in order to avoid the influence of secondary ionization process, the gas pressure should not be over 2000 Pa in the SST experiments. The effective ionization coefficients \((\alpha - \eta)/N\) measured were plotted in Figure 5 and critical electric field \((E/N)_{\text{lim}}\) were listed in Table 2. It could be found that the effect of different gas pressure at 500, 1000 and 2000 Pa on the results is not significant, even could be negligible within the range of the SST experimental condition. Comparing to the dielectric strength obtained by breakdown tests at high gas pressure, the insulation ability in 9.00\%C\(_4\)F\(_7\)N/91.00\%CO\(_2\) is weaker at low gas pressure obtained in SST experiment. However, due to the limitation of SST method, it is impossible to carry out the experiments at higher gas pressure over 2000 Pa.

According to Chachereau’s researches [10], [11], the different results of dielectric strength measured in SST experiment and breakdown test could be explained with the ion kinetics of C\(_4\)F\(_7\)N at different gas pressure. As reported, there are three different negative ions in C\(_4\)F\(_7\)N discharges, and all these three ions could have electron detachment process. While, at low gas pressure and small geometric distance, which is the SST experimental condition in this study, the electron detachment could be ignored. However, electron detachment could affect the discharge more strongly by forming a significant current from detached electrons when the gas pressure is high. Moreover, it should be noticed that the detachment of short-lived anion could only be observed at low pressure and the detaching anions do no contribute to the electric strength at high gas pressure. Then, the ion kinetics could have an effect on discharge process at different gas pressures. Therefore, the dielectric strength evaluated by the \((E/N)_{\text{lim}}\) obtained by SST experiments could be different from the results obtained in the breakdown test at high gas pressure.

**E. THE BOILING POINT IN C\(_4\) F\(_7\) N/CO\(_2\) MIXTURES**

The liquefaction temperature (i.e. boiling point) of the mixed gas at high pressure is an important indicator that cannot be ignored, when considering the actual operating conditions. In the high voltage applications such as GIL, the gas pressure of insulation gas is at least 0.3 MPa usually. According to the Antoine equation [20], [21], the relationship between the saturated vapor pressure \(p\) and the boiling point \(T\) could be
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since the electron detachment could be ignored at low gas pressure and small geometric distance.

The value of $(E/N)_{\text{lim}}$ in 13.13% C4F7N/86.87% CO2 is 292.44 Td, which is approximately 80% that of SF6. However, when the liquefaction temperatures of the gases are considered, the C4F7N/CO2 gas mixtures with 9.00% C4F7N contents, which has the critical electric field over 70% of SF6 gas, can be considered as potential candidate insulation gases to replace SF6 in the high voltage engineering field in the future.

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YUNXIANG LONG was born in Hubei, China, in 1992. He received the B.S. degree in physics from Wuhan University, China, in 2015, where he is currently pursuing the Ph.D. degree in particle physics and nuclear physics. His research interests focus on the insulation performance of alternative gases to SF6 gas.

LIPING GUO was born in Hubei, China, in 1968. He received the B.S. degree in physics from Jilin University, China, in 1989, the M.S. degree in condensed matter physics from the China Institute of Atomic Energy, in 1999, and the Ph.D. degree in particle physics and nuclear physics from the China Institute of Atomic Energy, in 2003. Since 2003, he has been a Professor with the School of Physics and Technology, Wuhan University. He is engaged in research on irradiation damage of nuclear materials and the insulation performance of gases.

CHENG CHEN was born in Hubei, China, in 1993. He received the B.S. degree in physics from the Dalian University of Technology, Liaoning, China, in 2015. He is currently pursuing the Ph.D. degree in physics with Wuhan University, China. His research interests focus on the calculation of gas insulation performance and the simulation of irradiation damage.

FANG LI was born in Hunan, China, in 1992. She received the B.S degree in physics from Jinan University, in 2017. She is currently pursuing the master’s degree with Wuhan University, focusing on the irradiation damage of nuclear materials and the insulation performance of gases.

WENJUN ZHOU (Senior Member, IEEE) was born in China, in July 1959. He received the Ph.D. degree from the Wuhan University of Hydraulic and Electrical Engineering, in 1990. He is currently a Professor with the School of Electrical Engineering and Automation, Wuhan University, China. He is a member of the High Voltage Committee of the Chinese Society of Electrical Engineering (CSEE), and the China Lightning Protection Standard Committee. His research interests include lightning protection and the diagnostic techniques for outdoor.

ZHENYU SHEN was born in Wuhan, China, in 1991. He received the B.S. degree in physics from Wuhan University, Wuhan, China, in 2013, where he is currently pursuing the Ph.D. degree in physics. His research interests focus on irradiation damage of nuclear materials and the insulation performance of gases.

YIHENG CHEN was born in Henan, China, in 1995. He received the B.S. degree in physics from Wuhan University, Wuhan, China, in 2017, where he is currently pursuing the master’s degree in particle physics and nuclear physics. His research interests focus on irradiation damage of nuclear materials and the insulation performance of gases.

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